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The symmetry of quasiperiodic crystals

R. Lifshitz*

Condensed Matter Physics 114-36, California Institute of Technology, Pasadena, CA 91125, USA

Abstract

Experimentally observed crystals range from periodic crystals, through incommensurately modulated crystals and composite crystals, to quasicrystals and even modulated quasicrystals. How does one characterize in a unified manner the symmetry of all these types of crystals? How does one classify all crystals according to their symmetry? These questions are answered through a review of the Fourier-space approach to crystal symmetry of Rokhsar, Wright, and Mermin. The notion of indistinguishability, which is central to the approach, is introduced and used as the basis for a generalization of the traditional space-group classification scheme, applicable to all types of crystals known to date.

1. Introduction

The discovery by Dan Shechtman [28] in 1982 of the first quasicrystal has launched an exciting reevaluation of our understanding of the nature of the crystalline state, one which is still far from being complete. It is clear that the old paradigm of a crystal as a periodic arrangement of identical unit cells can no longer hold. The Commission on Aperiodic Crystals of the International Union of Crystallography [35, page 928] has made this statement official in 1991 by giving a new definition to crystal:

“...by *crystal* we mean any solid having an essentially discrete diffraction diagram, and by *aperiodic crystal* we mean any crystal in which three-dimensional lattice periodicity can be considered to be absent.”

The new definition was intentionally made vague by the inclusion of the word “essentially”. It was meant only as a temporary working-definition until a better understanding of crystallinity emerges. We shall adopt it for the purpose of our discussion here, but we shall consider only those crystals whose density functions may be described as a superposition of a countable number of plane waves. These are crystals which ideally (disregarding such effects as thermal diffuse scattering) would produce diffraction

* e-mail: lifshitz@cco.caltech.edu

diagrams that may be expressed as sums of delta functions. We shall refer to such aperiodic crystals as *quasiperiodic crystals*.¹ All the experimentally observed crystals, whether modulated crystals, composite crystals, or quasicrystals – a distinction which will be made clear in Section 2 – belong to this category.

In this paper we shall be concerned with the symmetry of crystals. We shall see that one needs to introduce new ideas in order to capture the notion of symmetry in quasiperiodic crystals. The traditional theory of crystal symmetry, which was developed along the lines of the old paradigm, describes the symmetry of a crystal by its space group, the set of rigid motions in three-dimensional space – combinations of translations, rotations, and reflections – that leave the crystal invariant. Such a description is not valid for quasiperiodic crystals, not only because there are no longer any translations that leave the crystal invariant, but also because there are, in general, no rotations that leave the crystal invariant. Yet, quasiperiodic crystals clearly possess some kind of symmetry.

We shall describe here a generalization of the space-group classification scheme that does not rely on spatial periodicity and captures the new notion of symmetry in quasiperiodic crystals. This is accomplished by introducing the idea of *indistinguishability* and using it to redefine the concept of point-group symmetry. This reformulation of conventional crystallography is most naturally described in Fourier space, where it becomes a symmetry-based classification scheme for diffraction patterns consisting of sharp Bragg peaks in line with the new definition of crystal. When those diffraction patterns can be indexed by three integers, the general scheme reduces to the traditional space-group classification, but the same scheme works just as well for the diffraction patterns of quasiperiodic crystals, which require more than 3 integers for their indexing. Furthermore, we shall show that the scheme is applicable to all types of quasiperiodic crystals requiring only minor modifications for the cases of modulated crystals and composite crystals.

The idea of describing space groups in Fourier space was proposed over 30 years ago by Bienenstock and Ewald [1]. Only after the discovery of quasicrystals was it established as a practical and unified symmetry-classification scheme for periodic and quasiperiodic crystals by Rokhsar, Wright, and Mermin [24, 25], with later contributions by Rabson, Ho, Dräger, and Lifshitz².

There exists an alternative extension of the space-group classification scheme to quasiperiodic crystals, which will not be discussed here. It was developed by de Wolff, Janssen, and Janner [8, 32, 34] and was used to classify the symmetry of the simplest incommensurately modulated crystals even before the discovery of the first quasicrystal. Their approach treats quasiperiodic crystals as three-dimensional sections of structures periodic in a higher-dimensional “superspace”. The symmetry of the quasiperiodic

¹ Note that the new definition is sufficiently vague to allow even an ideal *aperiodic* crystal to have other terms in its Fourier spectrum in addition to a sum of delta functions. As defined here, the density of a *quasiperiodic* crystal is an *almost periodic function* of \mathbf{r} , according to Bohr [2].

² For a recent review of the Fourier-space approach see [15]. For a more detailed argument establishing the applicability of the approach to all types of quasiperiodic crystals see [10].

crystal is then given by the high-dimensional space group describing the set of rigid motions in superspace that leave invariant the high-dimensional structure. The superspace approach, which was originally developed for treating incommensurately modulated crystals, has since been extended to deal also with composite crystals [7, 30, 31, 33] and with quasicrystals [9]. In each case special measures need to be taken to accommodate the particular type of quasiperiodic crystal.

In Section 2 we introduce the different types of quasiperiodic crystals. In Section 3 we discuss the description of the symmetry of a given crystal, leading to the generalization of the space-group concept. We then show in Section 4 how this generalization is used as a basis for a symmetry classification scheme for all crystals. In Section 5 we discuss the phenomenon of extinctions, which has a straightforward explanation when viewed from the perspective of Fourier space. We finish in Section 6 with a selection of examples illustrating some of the ideas used in enumerating the possible symmetry classes of crystals, and outlining some of the problems that are yet to be solved.

2. The different types of quasiperiodic crystals

Although an official nomenclature has not yet been established, one clearly distinguishes between different kinds of quasiperiodic crystals: incommensurately modulated crystals, incommensurate composite crystals, and quasicrystals. We should clarify what we mean by these terms.³

The simplest quasiperiodic crystal is an *incommensurately-modulated periodic crystal*. It can be considered as a periodic basic (or average) structure which is perturbed in a periodic way (“modulated”), with the period of the perturbation incommensurate with the underlying periodicity of the basic structure. The nature of the perturbation is usually a periodic displacement of the atomic positions or a periodic variation in the occupation probability of the atomic sites. The diffraction diagrams of incommensurately modulated periodic crystals are characterized by having a subset of “main reflections” – Bragg peaks which are brighter than the others – that can be indexed by three integers and forms an ordinary commensurate reciprocal lattice. This lattice describes the periodicity of the average periodic structure. The weaker peaks, called “satellites”, describe the periodicity of the modulation.

Incommensurate composite crystals, also called intergrowth compounds,⁴ can be considered as composed of two or more interpenetrating subsystems with mutually incommensurate periodicities. Each subsystem when viewed independently is itself a crystal – in all known examples a periodic one – which is incommensurately modulated due to its interaction with the other subsystems. Examples of composite crystals are

³ See also van Smaalen’s recent review on quasiperiodic crystals [29]. Our definitions of the different kinds of quasiperiodic crystals differ from the ones given there in two ways: (1) Van Smaalen requires the average structure of a modulated crystal and the subsystems of a composite crystal to be periodic. (2) He defines quasicrystals as crystals with “non-crystallographic” point groups.

⁴ We follow here the nomenclature used by van Smaalen [31].

misfit layer structures which consist of alternating layers of the different subsystems where the periodicity within the layers of one subsystem is incommensurate with those of the other subsystems. There are also *channel-type structures* which possess one host structure containing channels in which the other subsystem(s) are accommodated. The diffraction diagrams of composite crystals are characterized by the existence of two or more subsets of main reflections, each forming a commensurate reciprocal lattice, caused by the average structure of one of the periodic subsystems, and a set of weak reflections caused by the modulations.

There are also quasiperiodic crystals for which a description in terms of a modulation of a basic structure or a composition of two or more substructures is either inappropriate or impossible. One frequently encountered model for such crystals is a quasiperiodic tiling such as the famous Penrose tiling. One fills space with “unit cells” or “tiles” in a way that maintains long-range order (and produces a discrete diffraction diagram) without being periodic. The observed icosahedral quasicrystals as well as the octagonal, decagonal, and dodecagonal quasicrystals fall under this category. There are also examples of quasiperiodic crystals with cubic symmetry [6], and possibly also examples with tetrahedral [4, 5] and with hexagonal [26] symmetry, that are neither modulated crystals nor composite crystals. We refer to all such quasiperiodic crystals as *quasicrystals*. We caution the reader that in most of the literature the term “quasicrystal” still refers *only* to quasiperiodic crystals which violate the “crystallographic restriction”, i.e. those which have an n -fold axis of symmetry with $n = 5$ or $n \geq 7$.

Finally, one can of course have *incommensurately modulated quasicrystals* [3, 14], where a quasiperiodic basic structure is incommensurately modulated. One can also, at least theoretically, construct composite crystals where the subsystems are themselves already quasiperiodic.

3. Symmetry description — The notion of indistinguishability

In trying to understand the symmetry of a quasiperiodic crystal it might be helpful to have in mind the two pictures shown in Fig. 1 – that of a typical diffraction pattern and that of a typical real-space model of a quasicrystal. The diffraction pattern immediately reveals a certain kind of symmetry. This is expressed by the set of (proper or improper) rotations, applied about the origin of wave-vector space, which leave invariant the *positions* of the Bragg peaks. We shall discuss this further in Section 3.1. But what exactly is the nature of the symmetry exhibited by the crystal itself (or by the tiling which represents it)? What do we mean when we say that a crystal has the symmetry of a certain rotation?

In the case of a periodic crystal we mean that the rotation leaves the density of the crystal invariant to within a translation. The densities of quasiperiodic crystals, however, in general possess no such symmetries. In fact, it is an easy exercise to show that if a two-dimensional crystal does contain more than a single point, about which an n -fold rotation ($n > 2$) brings it into perfect coincidence with

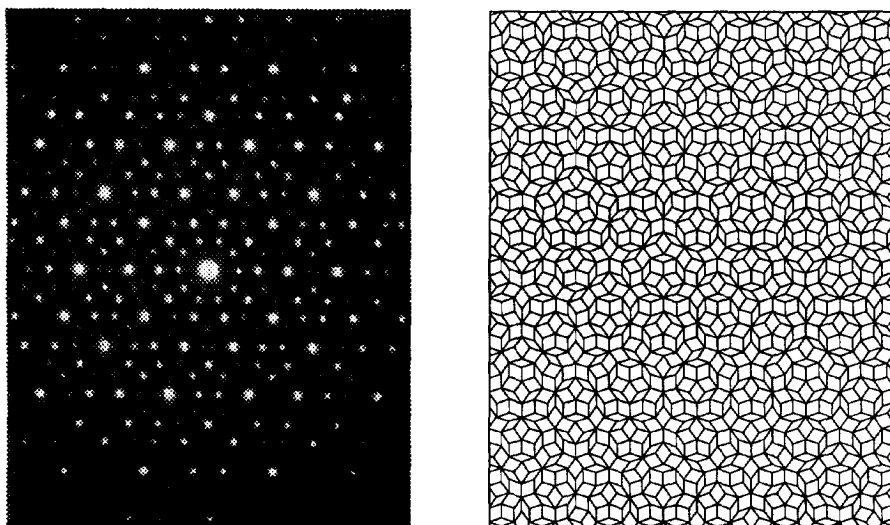


Fig. 1. On the left, a typical diffraction pattern of an icosahedral quasicrystal taken along an axis of 5-fold symmetry (source unknown). On the right, a section of the Penrose tiling, a model of a two-dimensional crystal with 10-fold symmetry.

itself, then the crystal is necessarily periodic. Nevertheless, certain rotations, when applied to a quasiperiodic crystal, take it into one which looks very much like the original unrotated crystal. This is because the two crystals contain the same statistical distribution of bounded substructures of arbitrary size. One observes that any bounded region in the rotated quasiperiodic crystal can be found in the unrotated crystal, but the larger the region the further away one has to look in order to find it.

Two densities that are statistically the same in this sense though not necessarily identical are called *indistinguishable*. The precise mathematical condition for densities ρ and ρ' to be indistinguishable is that they have the same positionally averaged n -point autocorrelation functions for all n :

$$\begin{aligned} & \lim_{V \rightarrow \infty} \frac{1}{V} \int d\mathbf{r} \rho(\mathbf{r}_1 - \mathbf{r}) \dots \rho(\mathbf{r}_n - \mathbf{r}) \\ &= \lim_{V \rightarrow \infty} \frac{1}{V} \int d\mathbf{r} \rho'(\mathbf{r}_1 - \mathbf{r}) \dots \rho'(\mathbf{r}_n - \mathbf{r}). \end{aligned} \quad (1)$$

The key to generalizing crystallography to include quasiperiodic crystals is thus to redefine the point group of the crystal to be the set of (proper or improper) rotations which leave it indistinguishable, i.e. which preserves all positionally averaged density autocorrelation functions. This redefinition reduces to the traditional one in the case of periodic crystals, and takes on a very simple form in Fourier space which is used to

give a concise description of the full space group symmetry of the crystal. This will be established in Section 3.2.

3.1. Lattices of wave vectors

Each Bragg peak in the diffraction pattern determines a wave vector \mathbf{k} at which the density of the crystal has a nonvanishing coefficient in its Fourier expansion,

$$\rho(\mathbf{r}) = \sum_{\mathbf{k} \in L} \rho(\mathbf{k}) e^{i\mathbf{k} \cdot \mathbf{r}}. \quad (2)$$

The (reciprocal) *lattice* L is defined as the set of all integral linear combinations of the wave vectors determined by the observed Bragg peaks. As so defined, L includes wave vectors at which the coefficients $\rho(\mathbf{k})$ are too weak to be detected in the diffraction experiment. As the resolution is improved more peaks may appear at larger wave vectors and in the quasiperiodic case, as seen in Fig. 1, also between already existing peaks. This is because quasiperiodic lattices are ‘dense’ in the mathematical sense – there is no requirement of minimal distance between wave vectors. The lattice L may also include wave vectors at which $\rho(\mathbf{k})$ is required to vanish by the symmetry of the crystal. Such wave vectors that are in the lattice but are missing from the Fourier expansion of the density are related to the phenomenon of extinctions which will be discussed in Section 5.

The lattices of all observed crystals may be expressed as integral linear combinations of a *finite* number of wave vectors. The minimum number D of vectors needed to generate the lattice is called its *rank* or its indexing dimension. A crystal is *periodic* if and only if the rank of its lattice is equal to the physical dimension d . Only then is the lattice a conventional ‘reciprocal lattice’ related in the familiar way to a lattice of real-space translations under which the periodic crystal is invariant.

We always use the term ‘lattice’ to refer to the rank- D lattice of wave vectors and not to any direct lattice of translations in ordinary (or higher-dimensional) real space. The lattice is an example of a \mathbb{Z} -module, which is defined as a vector space except that the scalars are restricted to be integers. Some crystallographers therefore like to call it *the* \mathbb{Z} -module or the Fourier module. We choose not to follow this nomenclature.

The point group G_L of the lattice (also called its holohedry) is the set of proper and improper rotations applied about the origin of Fourier space which leave the lattice invariant. We shall consider only finite⁵ subgroups of $O(3)$ and require that they contain the inversion because every lattice contains the negative of each of its vectors. The point group G of a crystal is necessarily a subgroup of the point group G_L of its lattice of wave vectors.

⁵Note that Piunikhin [20] has discovered that lattices of *finite* rank may have *infinite* point groups. Mermin [17] has given a simple construction of a two-dimensional lattice of rank 4 with infinite rotational symmetry.

3.2. The space group — A point group with phase functions

It is easily shown [15, p. 7] that the condition (1) for indistinguishability acquires a very simple form in Fourier space – the Fourier coefficients of two indistinguishable densities ρ and ρ' must be related by

$$\rho'(\mathbf{k}) = e^{2\pi i\chi(\mathbf{k})}\rho(\mathbf{k}), \quad (3)$$

where $\chi(\mathbf{k})$, called a *gauge function*, is linear modulo an integer over the lattice of wave vectors (i.e. $\chi(\mathbf{k}_1 + \mathbf{k}_2) \equiv \chi(\mathbf{k}_1) + \chi(\mathbf{k}_2)$ whenever \mathbf{k}_1 and \mathbf{k}_2 are in the lattice, where “ \equiv ” indicates equality modulo an integer).

In the periodic case one can show that $2\pi\chi(\mathbf{k})$ is necessarily of the form $\mathbf{k} \cdot \mathbf{d}$ for some constant vector \mathbf{d} independent of \mathbf{k} , so that $\rho'(\mathbf{r}) = \rho(\mathbf{r} + \mathbf{d})$ and indistinguishability reduces back to identity to within a translation. One can then combine point group operations with translations to recover the traditional space groups of periodic crystals, containing operations that leave the density *identical* to what it was. In the quasiperiodic case one must retain the general form of $\chi(\mathbf{k})$ which is defined only on the lattice and cannot be linearly extended to arbitrary \mathbf{k} .

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 $\rho(\mathbf{k})$:

$$\rho(g\mathbf{k}) = e^{2\pi i\Phi_g(\mathbf{k})}\rho(\mathbf{k}). \quad (4)$$

Since $\rho([gh]\mathbf{k}) = \rho(g[h\mathbf{k}])$, it follows directly from (4) that the set of phase functions associated with the elements of a point group G must satisfy the *group compatibility condition*:

$$\forall g, h \in G: \quad \Phi_{gh}(\mathbf{k}) \equiv \Phi_g(h\mathbf{k}) + \Phi_h(\mathbf{k}). \quad (5)$$

These constraints on the phase functions are the generalizations to quasiperiodic crystals of the Frobenius congruences in the traditional space group description of periodic crystals (see, for example, [27, p. 69]).

Putting all the pieces together, the *space group* of a crystal, which describes its symmetry, is given by:

- (i) A lattice of wave vectors L , invariant under some point group G_L , which only in the special case of periodic crystals is reciprocal to a lattice T of direct-space translations leaving the crystal invariant;
- (ii) A point group G , which is a subgroup of G_L , whose elements leave the density of the crystal indistinguishable, a criterion which in the case of periodic crystals reduces to identity to within a translation; and
- (iii) A set of phase functions $\Phi_g(\mathbf{k})$, one for every g in G , satisfying the group compatibility condition (5), which only in the periodic case may be given by a corresponding set \mathbf{d}_g of direct-space translations in the form $\mathbf{k} \cdot \mathbf{d}_g$.

We continue to call this a space group even though it is no longer a subgroup of the Euclidean group $E(3)$. Nevertheless, it may be given an algebraic structure of a group consisting of ordered pairs (g, Φ_g) as shown by Rabson et al. [21].

How does this description differ for the different types of quasiperiodic crystals introduced in Section 2? In essence there is no difference, in practice one proceeds as follows.

If no distinctions are made between strong and weak reflections in the diffraction pattern of a crystal then its symmetry is described by its space group as was just illustrated. This is the case for *periodic crystals* and for *quasicrystals*.

If the structure is a *modulated periodic crystal*, characterized by the existence of a single sublattice of main reflections, then one might want to supplement the specification of the space group by stating which of the possible sublattices of the full lattice contains the strong peaks. Identifying a particular sublattice of main reflections can be viewed as using a particular setting of the space group which emphasizes the existence of that sublattice. This is done by choosing a set of generating vectors for the full lattice of which the first three generate just the lattice of main reflections. Thus, the symmetry of the crystal is again described by its space group, given in an appropriate setting.

If the structure is a *composite crystal* with two or more sublattices of main reflections then one may supplement the space group description by specifying *all* of the sublattices that contain main reflections, i.e. by listing all the appropriate settings. The same procedure applies to the case of *modulated quasicrystals* or hypothetical *composite quasicrystals* with the only difference that one is looking for sublattices of main reflections whose rank is greater than 3.

4. Symmetry classification – Bravais classes and space group types

There are infinitely many crystal structures each of which has a space group describing its symmetry. The common symmetry properties of the different crystal structures become clear only after they are classified into properly chosen equivalence classes. We are concerned here with the classification of crystals into Bravais classes (Section 4.1), point groups, and space group types (Section 4.2).⁶

4.1. The Bravais class – A class of (reciprocal) lattices

Crystals are classified into Bravais classes according to their lattices of wave vectors. Two lattices are in the same *Bravais class* if one can interpolate between them with a sequence of lattices, all with the same point group and rank. Such an interpolation provides an isomorphism between the two lattices as additive abelian groups which also preserves the action of the point group. The relative intensities of the

⁶ Other classifications are also possible, see, for example, [27, Section 6.3].

Bragg peaks associated with the lattice wave vectors do not come into this definition of Bravais class, only the positions of the wave vectors in the lattice are important.

4.2. The space group type – A class of phase functions

The finer classification of crystals in a given Bravais class with a given point group is an organization of sets of phase functions into equivalence classes according to two criteria.

1. *Gauge equivalence.* Two sets of phase functions, Φ and Φ' , that describe indistinguishable densities ρ and ρ' , related by a gauge function χ , are associated with the same symmetry class. It follows from (3) and (4) that two such sets must be related by

$$\Phi'_g(\mathbf{k}) \equiv \Phi_g(\mathbf{k}) + \chi(g\mathbf{k} - \mathbf{k}) \quad (6)$$

for every g in the point group and every \mathbf{k} in the lattice. We call phase functions describing indistinguishable densities *gauge-equivalent* and Eq. 6, converting Φ into Φ' , a *gauge transformation*.

2. *Scale equivalence.* Two sets of phase functions, Φ and Φ' , are also counted as equivalent if there is a symmetry s of the lattice L , which is an automorphism of the point group $G = sGs^{-1}$, taking one set into the other

$$\Phi'_g(\mathbf{k}) \equiv \Phi_{sgs^{-1}}(s\mathbf{k}). \quad (7)$$

Operations s that are in the point group G of the crystal have precisely this property, but one can 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 sets of phase functions will not in general be gauge-equivalent. In the periodic case s can be an element of $O(3)$ (for example, a 90° rotation when G is a tetrahedral point group on a cubic lattice), or an element of $O(3)$ combined with a rescaling of the primitive lattice-generating vectors (for example, 90° rotations of an orthorhombic lattice). Here the distinct gauge-equivalence classes making up a single space group type are the different settings of that space group type. In the quasiperiodic case s can be an isotropic rescaling of the entire lattice (as in icosahedral quasicrystals), an isotropic rescaling of a sublattice (as in axial quasicrystals), or even independent rescalings of individual 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 called *scale-equivalent*.

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

5. Gauge-invariant phases – Screw axes, glide planes, and extinctions

It follows from the form of the gauge transformation (6) that if $g\mathbf{k} = \mathbf{k}$ then $\Phi_g(\mathbf{k})$ is independent of the choice of gauge. These gauge-invariant phases are directly related to the phenomenon of *extinctions*, for it follows directly from the definition (4) of the phase function that whenever $g\mathbf{k} = \mathbf{k}$, the Fourier coefficient $\rho(\mathbf{k})$ vanishes unless $\Phi_g(\mathbf{k})$ also vanishes (modulo an integer). Thus, the phase functions of a given space group type immediately determine the extinctions. It has been shown by Mermin [16] that there is no other generic reason for wave vectors in the lattice to be missing from the Fourier expansion of the density.

Extinctions have a somewhat different character when viewed from the perspective of Fourier space. Traditional crystallography starts with a direct lattice of translations, dual to this lattice is a lattice of wave vectors, and associated with each wave vector is a Bragg peak. Certain Bragg peaks may be missing from the diffraction pattern as a consequence of special symmetry elements of the space group – screw axes and glide planes. In Fourier-space crystallography one begins with the observed diffraction pattern and extends it by taking all integral linear combinations of observed wave vectors to form the lattice. Every lattice vector is a candidate for an additional Bragg peak unless the associated Fourier coefficient is required by the space group to vanish. The emphasis thus shifts from extinctions as missing Bragg peaks to extinctions as peaks that can never be added to the diffraction pattern no matter how much the resolution is improved.

One can easily show by using the group compatibility condition (5) that a phase function associated with a mirror m can only assume the values 0 and $\frac{1}{2}$ in the invariant plane of the mirror, and that a phase function associated with an n -fold rotation r can only assume the values j/n ($j = 0, 1, \dots, n - 1$) on the axis of rotation.

We call the plane of a mirror m a *glide plane* if $\Phi_m(\mathbf{k}) \not\equiv 0$ for any lattice vector in the plane. We call the axis of a rotation r a *screw axis* if $\Phi_r(\mathbf{k}) \not\equiv 0$ for any lattice vector on the axis. These definitions eliminate the need to associate extinctions with the interplay between rotations and translations which is peculiar to the periodic case.

6. Enumeration – Basic ideas and selected examples

We finish with a selection of examples illustrating some of the ideas used in enumerating the possible Bravais classes and space group types, and outlining some of the problems that are yet to be solved. In going through these examples we would like to emphasize the three-dimensional geometric nature of the enumeration procedures, as well as their generality in dealing with the different types of quasiperiodic crystals.

6.1. Bravais classes

Enumerating all Bravais classes of lattices of a given point group and rank can be a highly non-trivial task. Consider, for example, the case of two-dimensional N -fold

symmetric lattices whose rank is the smallest compatible with their point group (N is necessarily even because in two dimensions the 2-fold rotation, which takes every wave vector to its negative, is always a symmetry of the lattice). The simplest lattices are given by all integral linear combinations of N unit vectors separated in angle by $2\pi/N$ and are called *standard lattices*. Mermin et al. [19] showed that a mapping of the enumeration problem to the mathematical theory of cyclotomic integers (all integral linear combinations of the N th roots of unity) reveals a surprising result. All 2-dimensional N -fold symmetric lattices (of lowest rank) are equivalent to the standard lattices for all N from 4 to 44 and for $N = 48, 50, 60, 66, 70, 84$, and 90. For all other N there are also non-standard lattices and the number of Bravais classes into which they are classified can be enormous (for example 359 057 for $N = 128$). Though real crystals are not known to exist with axes of symmetry of such high order, one should not take lightly the statement, for example, that all 12-fold lattices of rank 4 belong to one Bravais class.

With this word of caution in mind we can go ahead and try to develop some systematic techniques for the enumeration of Bravais classes. One such technique is the “modular lattice method”. Simply stated it requires one to find a common sublattice for all lattices of a given rank and point group, and then to consider all the distinct ways of adding lattice points to construct the full lattice. This way of looking at all possible lattices corresponds, for example, to viewing each rank-3 cubic lattice as a simple cubic lattice with a basis. The significance of the method is in reducing the Bravais class counting to a finite procedure by having to consider only a finite number of modular lattices.

The modular lattice method was used by Rokhsar et al. [23] to enumerate the rank-6 icosahedral Bravais classes. It was also used by Mermin and Lifshitz [18] to enumerate the Bravais classes of the simplest *reducible* incommensurate lattices – all the three-dimensional rank-4 Bravais classes and all the three-dimensional rank-6 cubic and tetrahedral Bravais classes. By “reducible” we mean that these lattices can be expressed as integral linear combinations of vectors from a rank-3 sublattice, independently invariant under the point group, and vectors from an additional rank-1 (or in the cubic and tetrahedral case, rank-3) lattice. Such lattices are the simplest lattices that can be used to describe the symmetry of incommensurately modulated crystals or composite crystals because of the need to associate invariant rank-3 sublattices with the sets of main reflections.

It was shown that all these reducible Bravais classes, except for the rank-6 tetrahedral ones, are not only reducible but also *decomposable*, meaning that the additional rank-1 (or in the cubic case, rank-3) lattice is also independently invariant under the point group. This implies, for example, that the rank-6 cubic lattices are all sums of two of the well-known rank-3 cubic lattices (the primitive P , body centered I^* , and face centered F^* – the star indicating that the lattices are so centered in Fourier space). As such they are classified into six Bravais classes denoted by the two constituent rank-3 Bravais classes as: $P + P$, $I^* + I^*$, $F^* + F^*$, $P + F^*$, $P + I^*$, and $I^* + F^*$. There are three Bravais classes of rank-6 tetrahedral lattices which cannot be decomposed in

such a way since we know that there are no tetrahedral lattices of rank 3, yet they are $3 + 3$ reducible. Lattices in these three Bravais classes are most simply described as integral linear combinations of the six vectors $(1, \pm\alpha, 0)$, $(0, 1, \pm\alpha)$, and $(\pm\alpha, 0, 1)$ with primitive indexing, face-centered indexing (the sum of all integers even) and body-centered indexing (all integers of the same parity). When the irrational number α is equal to the golden mean these become the three Bravais classes of rank-6 icosahedral crystals.

Using a variant of the modular lattice idea, Lifshitz and Mermin [11, 12] have given a general proof that all hexagonal and trigonal lattices of arbitrary (but finite) rank are decomposable into only three types of periodic building blocks: rank-1 sublattices along the axis of 3- or 6-fold symmetry, rank-2 triangular sublattices in the plane perpendicular to the axis of rotation, and – only in the trigonal case – rank-3 rhombohedral sublattices. One can then use this property of the lattices to enumerate all hexagonal and trigonal Bravais classes of arbitrary rank- n by simply considering all the distinct ways of putting these building blocks together such that the rank adds up to n .

A more general question to which we still do not have an answer is how to determine the set of indecomposable Bravais classes for any given point group. We have shown, for example, that all three-dimensional rank-4 lattices are decomposable. Is this an indication that in the orthorhombic system, as in the trigonal-hexagonal system, all indecomposable lattices are of rank 3 or less, or are there indecomposable orthorhombic lattice of rank greater than 4? This question is related to finding all the *indecomposable integral representations* of a given group, for which, as far as we know, no simple and general answer exists (as in the cases of complex or real representations).

6.2. Gauge-equivalence classes

When the lattice of wave vectors has finite rank the procedure for determining the possible phase functions is straightforward because they need be specified only by their values at the lattice-generating vectors (due to their linearity), and only for a set of elements g sufficient to generate the point group G (due to the group compatibility condition). These values are constrained by applying the group compatibility condition to the point group generating relations. By making a judicious choice of gauge one can simplify the calculation from the start by setting many of the unknown phases to zero, extracting a unique representative for each class of gauge-equivalent phase functions. As an example [15], all the icosahedral space groups may be specified by a single phase function even though the two icosahedral point groups contain 60 and 120 elements.

When the lattice is decomposable into a sum of sublattices of lower rank, each independently invariant under the lattice point group, it is not necessary to recompute the possible gauge-equivalence classes for the high-rank lattice. One only needs to consider all the different combinations of the gauge-equivalence classes already classified for the lower-rank sublattices. This is possible because the group compatibility condition (5)

acts independently in each invariant sublattice and the choice of gauge is independent in each invariant sublattice. This idea was greatly exploited by Lifshitz and Mermin [11,12].

6.3. Scale-equivalence classes

The remaining part of the space-group classification is merely a matter of simplifying the bookkeeping by grouping together different gauge-equivalence classes which are scale-equivalent. Although the grounds for this further identification are stated quite precisely, whether one chooses to make it or not can be a matter of convention. In point groups which lack-inversion symmetry, for example, the inversion is still a symmetry of the lattice and as such it can be used to relate gauge-inequivalent phase functions. In the periodic case, one normally chooses not to make this identification (because the inversion is not sense preserving), counting pairs of right-handed and left-handed gauge-equivalence classes as distinct space group types.

When dealing with incommensurately modulated crystals or composite crystals one may wish to constrain the rescaling operations s to those which leave invariant the sublattice(s) of main reflections. In this way one can directly obtain the different settings of the space groups which are appropriate for the description of such crystals. By first focusing only on the gauge-equivalence classes of phase functions, we give the results of the non-trivial 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 bookkeeping question of which classes to further identify through scale-equivalence we retain the freedom to use whatever transformations are appropriate to the material of interest. In such a way we recover both the space groups and their settings which may be used to emphasize particular sublattices of main reflections, making straightforward the treatment of materials even when they fail to fit neatly into any particular conventional category (modulated crystals, composite crystals, and quasicrystals) and allowing for a unified description of materials that might interpolate between the different categories.

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