Siegbert Schmid · Ray L. Withers
Ron Lifshitz *Editors*

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Preface

Aperiodic Crystals collects 37 selected papers from the scientific contributions presented at the Seventh International Conference on Aperiodic Crystals, *Aperiodic 2012*, held in Cairns, Australia from the 2nd to the 7th of September, 2012 and organized under the auspices of the Commission on Aperiodic Crystals of the International Union of Crystallography (IUCr). It followed Aperiodic'94 (Les Diablerets, Switzerland), Aperiodic'97 (Alpe d'Huez, France), Aperiodic 2000 (Nijmegen, The Netherlands), Aperiodic 2003 (Belo Horizonte, Brazil), Aperiodic 2006 (Zao, Japan) and Aperiodic 2009 (Liverpool, U.K.). The *Aperiodic* series of conferences in turn followed on four earlier conferences held under the title of Modulated Structures, Polytypes and Quasicrystals (MOSPOQ). The eighth conference in the *Aperiodic* series will be held in Prague in 2015.

The program was wonderfully diverse, covering a wide range of topics including: the mathematics of aperiodic long-range order and the fascinating types of tilings resulting from it; the synthesis, growth and stability of metallic aperiodic crystals and related complex metallic alloys; new methods and associated structural characterisation studies of aperiodic crystals; theoretical and experimental studies of the electronic, magnetic and other physical properties of aperiodic crystals; partial order, correlated disorder, and structured diffuse scattering; modulated structures, quasicrystals and approximants; soft-matter quasicrystals, and aperiodic ordering in bio-molecules and proteins; the dynamics of aperiodic crystals; as well as aperiodic surfaces, thin films and adsorbates. This impressive diversity in subject matter is well reflected in the contributions to this volume.

The conference was attended by more than 110 delegates from 23 different countries, including Dan Shechtman from Israel, Laureate of the 2011 Nobel Prize in Chemistry. Prof. Shechtman delivered a special celebratory Nobel lecture on the 30th anniversary year of his pioneering electron-diffraction characterization of the first quasicrystal on April 8, 1982. The introductory tutorial talk by Ted Janssen highlighted the fact that we also celebrated a half a century of work on aperiodic crystals, which could be considered as dating back to the pioneering work of Pim de Wolff on γ-Na_2CO_3 around 1962 and published in 1964. Presentations at the conference included 3 invited introductory tutorials, 11 invited talks, 46 contributed talks, and 43 poster presentations, all discussing state-of-the-art research in this fascinating field of scientific endeavour. What we know and what we still don't know about aperiodic order was carefully examined and hotly debated throughout this conference.

We would like to thank all the participants for coming the very long way to Australia as well as for their enthusiastic and considered contributions to, and participation in, the conference. Special thanks are due to the International Program Committee for their work in the organisation of the conference program and to the members of the Local Organizing Committee for making Aperiodic 2012 the very successful and highly stimulating meeting it was. We would also like to thank the wonderful staff from Springer for their help in the production of this volume. Finally, we gratefully acknowledge financial and other support from our sponsors.

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Chapter 1
A Brief History of Aperiodic Crystals: 1962–2012

T. Janssen

Abstract About 50 years ago, it was shown that there are solid state systems with perfect order but without lattice periodicity. These systems were called crystalline phases because of the order and incommensurate because of the lack of periodicity. They formed a challenge for crystallographers and physicists to understand the structure, the physical properties and the reason for their appearance. Later other classes of this type were found (occupation modulated crystals, incommensurate magnetic systems, incommensurate composites), the most important one being that of quasicrystals. The discovery of the latter class in 1982 caused a huge increase in interest. The first conferences on this new type of materials were called Modulated Crystals, later polytypes and quasicrystals were included in the title MOSPOQ. Nowadays these conferences continue under the name Aperiodic (Crystals). The field has become very active worldwide, and our insight into structure and properties has increased impressively. A brief sketch of the development of the field is given in this chapter.

1.1 History

On April 21st, 1912, Friedrich, Knipping and Laue [1] followed a proposal by Max Laue (after 1913 von Laue) and performed an experiment throwing X-rays onto a crystal of copper sulfate [2]. They found the sharp diffraction spots foreseen by Laue showing that the crystal has lattice periodicity. Shortly afterwards the Braggs developed the fundamental techniques of crystallography. For half a century the paradigm was that ideal crystals are built of identical unit cells. Later the International Union of Crystallography incorporated this property into the definition of a 'crystal'. This idea remained intact for half a century. Then systems with also sharp diffraction spots but without lattice periodicity were found. The idea to consider these materials also as crystals reached the larger crystallographic community much later, after Dan Shechtman had discovered quasicrystals on April 8th, 1982. The first aperiodic crystals, however, were discovered earlier, but this did not attract so much attention.

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It is more difficult to state when exactly the first aperiodic crystal was found. In 1960, a spin wave was found with a period that did not fit the periodicity of the underlying lattice [3], but one did not observe an effect on the positions of the atoms. In 1963, satellites were seen in $NaNO_2$ [4], next to the main reflections. These were interpreted as micro-domains, in a very small (1.5 degree centigrade) temperature interval, around the ferroelectric phase transition and there was no discussion of the incommensurate character. But in 1964 the first incommensurately modulated structure was found. So there is some reason to consider 1962 (between 1960 and 1964) as the beginning of the field.

1.1.1 Incommensurate Modulated Phases and Composites

In 1964, Pim de Wolff and collaborators [5] found satellite peaks in the γ-phase of anhydrous Na_2CO_3: next to the main reflections of the monoclinic basic structure there were peaks at positions $h\mathbf{a}^* + k\mathbf{b}^* + \ell c^* + m(\alpha\mathbf{a}^* + \beta\mathbf{b}^*)$. First they were found in powder, later also in a single crystal. The interpretation was a periodic displacement of the atoms, with wave vector $\mathbf{q}$ which has irrational indices α and β with respect to the reciprocal lattice vectors $\mathbf{a}^*$ and $\mathbf{b}^*$ of the basic structure. The conclusion is that the structure is not lattice periodic. Mathematically, it is a quasiperiodic structure, but to stress the aperiodicity these phases were called *incommensurately modulated phases*. Soon other examples of such structures followed, e.g. thiourea and K_2SeO_4, to mention two examples which were studied extensively. For such structures the positions of the atoms can be given as

$$\mathbf{r}_{\mathbf{n}j} = \mathbf{n} + \mathbf{r}_j + \mathbf{u}_j\big(\mathbf{q}.(\mathbf{n} + \mathbf{r}_j)\big), \tag{1.1}$$

where $\mathbf{n}$ are the lattice points of the lattice periodic basic structure, $\mathbf{r}_j$ the position of the jth atom in the unit cell, $\mathbf{u}$ a periodic function with period 1. Later, also structures with 2 or more modulation vectors $\mathbf{q}$ were found. The diffraction spots are at positions

$$\mathbf{H} = h\mathbf{a}^* + k\mathbf{b}^* + \ell\mathbf{c}^* + \sum_{s=1}^{d} m_s\mathbf{q}_s. \tag{1.2}$$

d is the number of independent modulation wave vectors. Main reflections are the peaks with $m_s = 0$. Because main reflections are mapped onto main reflections, the symmetry of the diffraction must be one of the three-dimensional crystallographic point groups.

A new type of aperiodic crystal was found in 1975. Tetrathiafulvalene (TTF) pentaiodide has a subsystem consisting of TTF molecules with as second subsystem iodide in channels, the compound having composition $TTF_7I_{5-\delta}$ [6]. The basic structure of the TTF system is C-centered monoclinic, that of the iodine system A-centered monoclinic, but the lattice constants are incommensurate. Therefore, one needs more than 3 basis vectors to index the diffraction spots with integers: one additional vector is needed. A second example is $Hg_{3-\delta}AsF_6$ [7], consisting of 3 subsystems: the AsF_6 host lattice, and two systems of mercury chains, one in the a-direction, and one in the b-direction. Each subsystem has a basic structure with 3 basis vectors for their reciprocal lattices. Nevertheless, instead of nine, one only needs four basis vectors for indexing the diffraction peaks with integers. Such structures with mutually incommensurate subsystems are called incommensurate composites. Since these first examples many other have been found. A particular class is that of misfit structures, layered structures for which the layers do not have the same translation symmetry, not even for the basic structure one has to consider, because generally the layers are modulated by the interaction with the other layers.

The diffraction pattern shows reflections common to several subsystems, reflections corresponding to main peaks for one of the subsystems and summation reflections corresponding to modulations of one subsystem caused by the interaction with other subsystems. The symmetry of the diffraction pattern consists of all orthogonal transformations mapping each spot to another of the same intensity. Because subsystems may be mapped on each other, the crystallographic condition no longer holds: in principle, the symmetry may contain non-crystallographic elements, i.e. elements which are impossible for a three-dimensional lattice periodic structure, such as a five-fold rotation. However, such symmetries have not been observed for composites.

1.1.2 Aperiodic Tilings and Quasicrystals

Non-crystallographic symmetries play a role in mathematical constructions, aperiodic tilings of the plane. The best known example is the Penrose tiling, a tiling of the plane with copies of two different tiles, without gaps or overlaps. One realization of a Penrose tiling is by means of rhombs, one with an angle of 36° and with an angle of 72°. An early overview was given by Martin Gardiner in the January 1977 issue of *Scientific American*. Crystallographers played with the idea that there could be crystals with a comparable structure. Alan Mackay [8] showed experimentally that the diffraction pattern has ten-fold symmetry. Later this could be proven mathematically. However, such structures, aperiodic and quasiperiodic, were not known by then.

Therefore, it was a big surprise when Dan Shechtman found real structures with sharp diffraction peaks and ten-fold symmetry in the diffraction pattern. He studied rapidly cooled AlMn particles and observed this phenomenon. Unfortunately, the referees were not aware of the existence of tilings with these properties, or they did not believe that this could happen in nature and did not accept the explanations for the findings. However, other explanations could all be proven to be false. It took two and a half years before the results could be published [9]. The new material did not only show ten-fold symmetry in the diffraction, but even the symmetry of an icosahedron. It became known as a quasicrystal. This discovery can be considered as the most important event in the history of aperiodic crystals.

Mackay already had used the term ‘quasi-lattice’ for the diffraction pattern of the Penrose tiling. For the new material Levine and Steinhardt proposed the term ‘quasicrystal’. It is supposed to mean ‘quasiperiodic crystal’, but the latter term is actually too broad. First, according to the original mathematical definition, any periodic function is also quasiperiodic; and second, there are aperiodic and quasiperiodic structures that are usually not considered to be quasicrystals, like the incommensurate phases. Actually, there is not yet a consensus about the term ‘quasicrystal’. ‘Aperiodic crystal’ is the general term for a structure with sharp diffraction peaks that is quasiperiodic and not lattice periodic.

After the discovery of the quasicrystalline structure in AlMn, new examples were rapidly found. Next to the icosahedral quasicrystals, there are the decagonal quasicrystals with quasiperiodicity in planes perpendicular to an axis along which the crystal is periodic. Also new classes of icosahedral quasicrystals were discovered which turned out to be of better quality and stable (the very small AlMn quasicrystals are unstable). Several families of ternary alloys (AlCuFe, AlMnPd, etc.) and binary alloys (e.g. YbCd) were developed. The larger size and higher quality were essential for the study of crystallographic and physical properties.

1.1.3 Incommensurate Magnetic Structures

As mentioned before, aperiodicity in crystals was found as an incommensurate spin wave in $MnAu_2$. There no influence of the spin wave on the crystal structure was reported. Such an interaction was found in chromium [10]. Below the Curie temperature, satellites are observed with a modulation wave vector equal to the spin wave vector. The coupling between spin and lattice causes a modulation, a mechanism comparable to the coupling between charge density waves and the lattice, leading to a modulation of the latter. Analogously to the displacive modulation (Eq. (1.1)), the spin wave at discrete positions may be written as

$$\mathbf{S}(\mathbf{n}, j) = \sum_{\mathbf{H}} \hat{\mathbf{S}}(\mathbf{H}) \exp\big(i\mathbf{H}.(\mathbf{n} + \mathbf{r}_j)\big). \tag{1.3}$$

Whereas displacive modulations are usually transversal or longitudinal, spin structures may show complicated spiral structures, especially in systems with rare-earth elements.

1.2 Superspace Treatment

A quasiperiodic function $f(x)$ is a function that is the restriction of a function g, periodic in each of its n variables, to a line in the n-dimensional space: $f(x) = g(\alpha_1 x, \ldots, \alpha_n x)$, where the numbers α_i are irrational. Notice that for $n = 1$ the function is periodic: a periodic function is also quasiperiodic. It may be shown that the projection of the Fourier transform of $f(x)$ on the line consists of sharp peaks at positions $q = \sum_{i=1}^{n} m_i q_i$. This corresponds to the observation by de Wolff that the peaks for γ-Na_2CO_3 can be seen as the projection points of a regular lattice in four dimensions.

The diffraction spots of a quasiperiodic crystal can be labeled with n indices. If $n = 3$ (or the dimension of the crystal which could also be 1 or 2), the crystal has lattice periodicity and the spots belong to the reciprocal lattice. In general, the spots are given as points on a *Fourier module*: linear combinations of n basis vectors $\mathbf{a}_i^*$. The number n is the *rank* of the Fourier module. If the orthogonal 3-dimensional transformation R leaves the pattern (inclusive intensities) invariant, one has

$$R\mathbf{a}_i^* = \sum_{j=1}^{n} \Gamma_{ij}(R)\mathbf{a}_j^*. \tag{1.4}$$

Because there are only a finite number of peaks above a certain intensity around the origin, the matrices $\Gamma(R)$ form a finite group, and group theory then tells us that this group on another basis consists of orthogonal matrices. Because the pattern in 3 dimensions is left invariant, the elements R correspond to pairs (R_E, R_I) of orthogonal transformations, in 3 and $(n-3)$ dimensions, respectively. The n-dimensional group then leaves an n-dimensional (reciprocal) lattice invariant, and its direct lattice as well. This is the general idea of constructing a periodic n-dimensional structure for which the restriction to 3 dimensions gives the physical, aperiodic crystal. An alternative way is the following. Suppose the aperiodic crystal has a density $\rho(\mathbf{r})$. Its Fourier component is non-zero only for points of the Fourier module, and these correspond to points of the n-dimensional reciprocal lattice. Then one can construct a lattice periodic function in n-dimensions

$$\rho(\mathbf{r}) = \sum_{\mathbf{H}} \hat{\rho}(\mathbf{H}) \exp(i\mathbf{H}.\mathbf{r}) \rightarrow \rho(\mathbf{r}, \mathbf{r}_I) = \sum_{\mathbf{H}} \hat{\rho}(\mathbf{H}) \exp\big(i(\mathbf{H}.\mathbf{r} + \mathbf{H}_I.\mathbf{r}_I)\big). \tag{1.5}$$

In the case of point atoms, the function $\rho(\mathbf{r}_E, \mathbf{r}_I)$ is restricted to $(n-3)$-dimensional hypersurfaces, called *atomic surfaces*. These may extend to infinity or be of finite volume, when they are disjunct. It is the goal of structure determination to find position and shape of these atomic surfaces. On them an occupation function may be defined which determines the probability of finding a certain chemical species there.

Because the projection of the lattice on internal space is a dense set, the energy of every 3-plane through a point of this set is the same, and this corresponds to a global translation along internal space. In many cases, local shifts do not cost much energy. Statically, this corresponds to phason disorder, dynamically to phason excitations (either jumps or collective motions). At finite temperature, there will be phason disorder which shows itself in points outside the atomic surfaces which may occur with a certain probability. The disorder could contribute to the entropy of the system, and in turn this might influence the balance between periodic and aperiodic structures.

Since the function $\rho(\mathbf{r}_E, \mathbf{r}_I)$ is lattice periodic, its symmetry group is an n-dimensional space group, a *superspace group*. An element g is a combination of a separable orthogonal transformation (R_E, R_I), where the two components are orthogonal transformations in physical and internal space, respectively, and a pair of transformations $(\mathbf{v}_E, \mathbf{v}_I)$ of translations in these 2 subspaces. Its action on $\rho(\mathbf{r}_E, \mathbf{r}_I)$ is given by

$$T_g\rho(\mathbf{r}_E, \mathbf{r}_I) = \rho\big(R_E^{-1}(\mathbf{r}_E - \mathbf{v}_E), R_I^{-1}(\mathbf{r}_I - \mathbf{v}_I)\big) \tag{1.6}$$

and on its Fourier transform by

$$T_g\hat{\rho}(\mathbf{k}) = \exp\big(i(\mathbf{k}_E.\mathbf{v}_E + \mathbf{k}_I.\mathbf{v}_I)\big)\hat{\rho}\big(R_E^{-1}\mathbf{k}\big). \tag{1.7}$$

This formula gives the systematic extinctions associated with the superspace group: if $R\mathbf{k} = \mathbf{k}$, then $\hat{\rho}(\mathbf{k}) = 0$, unless the argument of exp is a multiple of 2π.

Let us consider this procedure for the various families.

1.2.1 Incommensurate Modulated Phases and Composites

De Wolff [11] noticed that the positions of the diffraction pattern (Eq. (1.2)) for $d = 1$ can be seen as the projection of a reciprocal lattice in 4 dimensions. As 4th dimension one can consider the phase of the modulation wave (Eq. (1.1)). Then one gets

$$\mathbf{r}_{\mathbf{n}j}(\phi) = \mathbf{n} + \mathbf{r}_j + \mathbf{u}_j\big(\mathbf{q}.(\mathbf{n} + \mathbf{r}_j) + \phi\big), \tag{1.8}$$

which is equivalent to the expression to obtain a quasiperiodic function from a periodic function in a higher-dimensional space. The symmetry of the pattern (1.8) is a four-dimensional space group. Such space groups had been used by Janner and Janssen for the study of space-time symmetries of electrodynamic systems. Then the phase ϕ is taken over by the time t. Together the approach could be generalized to more general modulated phases, and later to general quasiperiodic crystals. This has been dealt with in the beginning of this section (Eq. (1.5)).

For incommensurate composites each subsystem (ν) has a lattice periodic basic structure with a reciprocal lattice on a basis $\mathbf{a}_i^{\nu *}$. A basis for the Fourier module, the set generated by all diffraction spot positions, is given by $\mathbf{a}_i^*$ $(n = 1, \ldots, n)$). The former can be expressed in the latter by $\mathbf{a}_i^{\nu *} = \sum_j Z_{ij}^{\nu}\mathbf{a}_j^*$. The basis $\mathbf{a}_i^*$ can be embedded into an n-dimensional space, and consequently the reciprocal and the direct lattice of each subsystem is embedded in an n-dimensional space. Each subsystem is modulated by the interaction with the other subsystems. This means that the main peaks of one may coincide with satellite peaks of other subsystems. In addition, there may be other modulations as well, which would increase the dimension of the superspace, but we shall disregard that possibility here.

1.2.2 Aperiodic Tilings and Quasicrystals

With some effort it is possible to index the diffraction pattern made by Mackay [8]. It has rank four, the symmetry group is the group $\bar{5}$m corresponding to a finite group of integer matrices, for which one can determine the invariant lattice in 4 dimensions and the unit cell. The problem is the determination of the atomic surfaces in this unit cell. It turns out that one has 4 different and disjunct atomic surfaces, all pentagons, of different size and orientation.

This illustrates the problems for the description of quasicrystals. The trivial part is the determination of the unit cell (or the asymmetric unit cell). This follows easily from the positions of the diffraction spots and the symmetry of the pattern. For decagonal (and octagonal or dodecagonal) quasicrystals the dimension of the superspace is five, for icosahedral quasicrystals six. Like for the Penrose tiling, the atomic surfaces here are disjunct. So one has to determine the position of these atomic surfaces and their shape. Moreover, there may be chemical order or disorder on the atomic surfaces. In the determination, the knowledge of the superspace group may help. Moreover, there is a closeness condition. This means the following. Two atomic surfaces with nearby positions in physical space do not overlap in projection on internal space. But if one changes the position of the n-dimensional crystal in internal space and an intersection point of an atomic surface with physical space vanishes, a new intersection point on another atomic surface should appear. This means that the projection of a border of an atomic surface should coincide with that of another atomic surface. This is the closeness condition which poses limitations on the shape of the atomic surfaces. Another helpful fact can be the existence of an approximant, a lattice periodic structure with similar chemical composition and similar local ordering. There are a number of structure models for ternary systems. Using the knowledge of the structure of an approximant, one has been able to determine the structure of the binary icosahedral YbCd [12].

1.2.3 *Incommensurate Magnetic Structures*

The description of incommensurate magnetic structures (Eq. (1.3)) and their effect on the nuclear structure in superspace follows the same lines. Both the spin waves and modulation waves may be embedded in superspace. The superspace group of the nuclear structure consists of all elements leaving it invariant. For the spin system one may introduce the time reversal operator θ. Then the action of the combination of a superspace group element g and θ on the spin wave in superspace is

$$T_g\theta\mathbf{S}(\mathbf{r}_E, \mathbf{r}_I) = -\mathrm{Det}(R_E)\mathbf{S}\big(g^{-1}(\mathbf{r}_E, \mathbf{r}_I)\big), \tag{1.9}$$

and for g itself the same expression holds without the minus-sign. The group of all elements g and $g\theta$ which leave $\mathbf{S}$ invariant is the magnetic superspace group. This group and the corresponding structure have been determined for several aperiodic magnetic structures such as chromium, rare-earth compounds and multiferroics [13–15].

1.3 Phase Transitions

As in conventional 3-dimensional crystals, aperiodic crystals may show phase transitions in the composition–pressure–temperature space. However, the variety is bigger for aperiodic systems. In the first place, one has to distinguish transitions where the dimension of superspace does not change, and those where there is a difference in dimension at the phase transition. An example of the former, is a transition to a superstructure, for example, a change in centering. If the transition is 2nd order, the order parameter is related to irreducible representations of the high-symmetry group, and one can apply Landau's theory of phase transitions.

Phase transitions where the dimension changes, are more typical for aperiodic crystals. Examples are the transition from an unmodulated to a modulated structure. These are often of 2nd order and related to a soft mode, a vibration mode becoming unstable at the transition. The appearing modulated structure may be described using irreducible representations of the symmetry of the unmodulated phase, at least near the phase transition. There is a relatively simple connection between such irreducible representations and the superspace group of the modulated phase [16]. Also modulations of quasicrystals may be described in this way, e.g. the icosahedral modulation of an icosahedral quasicrystal is a transition from a six- to a 12-dimensional structure.

For incommensurate composites the phase transition may correspond to a change in the relationship between the subsystems. Examples of such phase transitions have been observed in nonadecane-urea [17]. In the phase diagram, one finds structures with rank 3, 4 and 5.

1.4 Conclusion

In 50 years the field of aperiodic crystals has grown to a rich and important topic. There is a very large variety of systems, and these are interesting from various points of view. The number of systems is large, and so is the total amount of such crystals, because many minerals belong to this class of materials. Although many mathematical, physical and chemical questions have been answered, there still remain many fundamental open questions concerning the origin of aperiodic order, the growth of the order, the reason for the stability, and the character of elementary excitations. Finally, the question remains in how far this class of materials may lead to new applications.

Additional information "http://www.janssenweb.net/ted/janssen.htm" and the book "Aperiodic Crystals" by Ted Janssen, Gervais Chapuis and Marc de Boissieu, Oxford University Press 2007.

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Chapter 2
Squiral Diffraction

Uwe Grimm and Michael Baake

Abstract The Thue–Morse system is a paradigm of singular continuous diffraction in one dimension. Here, we consider a planar generalisation, constructed by a bijective block substitution rule, which is locally equivalent to the squiral inflation rule. For balanced weights, its diffraction is purely singular continuous. The diffraction measure is a two-dimensional Riesz product that can be calculated explicitly.

2.1 Introduction

The diffraction of (fully) periodic systems and of aperiodic structures based on cut and project sets (or model sets) is well understood; see [4, 5] and references therein. These systems (in the case of model sets under suitable assumptions on the window) are pure point diffractive, and the diffraction can be calculated explicitly.

The picture changes for structures with continuous diffraction. Not much is known in general, in particular for the case of singular continuous diffraction, even though both absolutely and singular continuous diffraction show up in real systems [13, 14]. The paradigm of singular continuous diffraction is the Thue–Morse chain, which in its balanced form (constructed via the primitive inflation rule $1 \mapsto 1\bar{1}$, $\bar{1} \mapsto \bar{1}1$ with weights 1 and $\bar{1} = -1$, say) shows purely singular continuous diffraction. This was shown by Kakutani [10], see also [1], and the result can be extended to an entire family of generalised Thue–Morse sequences [3].

Here, we describe a two-dimensional system which, in its balanced form, has purely singular continuous diffraction. For mathematical details, we refer to [6]. Again, it is possible to obtain an explicit formula for the diffraction measure in terms of a Riesz product [12, Sect. 1.3], with convergence in the vague topology.

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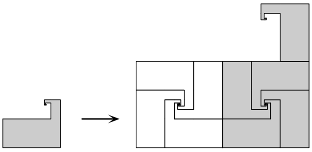

Fig. 2.1 The primitive inflation rule for the squiral tiling of the Euclidean plane

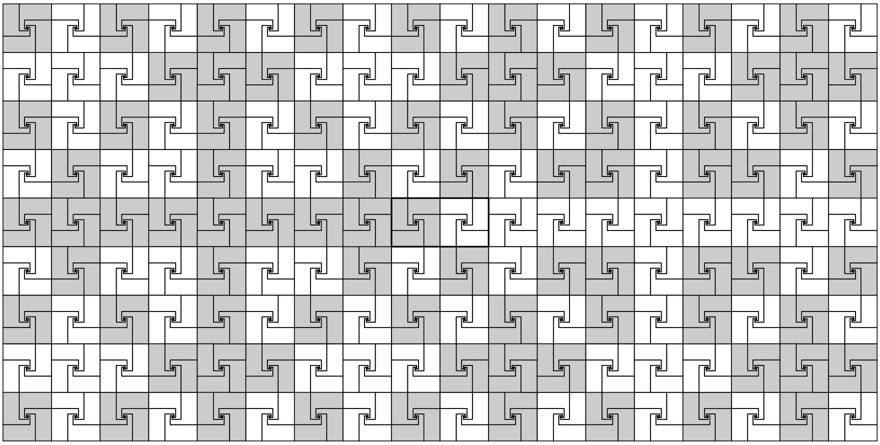

Fig. 2.2 Patch of the squiral tiling obtained by two inflation steps from the central seed

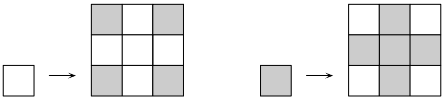

Fig. 2.3 Equivalent block inflation rule for the squiral tiling of Fig. 2.2

2.2 The Squiral Block Inflation

The squiral tiling (a name that comprises 'square' and 'spiral') was introduced in [8, Fig. 10.1.4] as an example of an inflation tiling with prototiles comprising infinitely many edges. The inflation rule is shown in Fig. 2.1; it is compatible with reflection symmetry, so that the reflected prototile is inflated accordingly.

A patch of the tiling is shown in Fig. 2.2. Clearly, the tiling consists of a two-colouring of the square lattice, with each square comprising four squiral tiles of the same chirality. The two-colouring can be obtained by the simple block inflation rule shown in Fig. 2.3, which is bijective in the sense of [11]. Again, the rule is compatible with colour exchange. The corresponding hull has D_4 symmetry, and also contains an element with exact individual D_4 symmetry; see [6] for details and an illustration.

Due to the dihedral symmetry of the inflation tiling, it suffices to consider a tiling of the positive quadrant. Using the lower left point of the square as the reference point, the induced block inflation ρ produces a two-cycle of configurations v and

ρv. They satisfy, for all $m, n \geq 0$ and $0 \leq r, s \leq 2$, the fixed point equations

$$(\rho v)_{3m+r,3n+s} = \begin{cases} \overline{v}_{m,n}, & \text{if } r \equiv s \equiv 0 \bmod 2, \\ v_{m,n}, & \text{otherwise.} \end{cases} \tag{2.1}$$

2.3 Autocorrelation and Diffraction Measure

For a fixed point tiling under ρ^2, we mark each (coloured) square by a point at its lower left corner $z \in \mathbb{Z}^2$. For the balanced version, each point carries a weight $w_z = 1$ (for white) or $w_z = \overline{1} = -1$ (for grey). Consider the corresponding Dirac comb

$$\omega = w\delta_{\mathbb{Z}^2} = \sum_{z \in \mathbb{Z}^2} w_z \delta_z, \tag{2.2}$$

which also is a special decoration of the original squiral tiling. Following the approach pioneered by Hof [9], the natural autocorrelation measure γ of ω is defined as

$$\gamma = \omega \circledast \tilde{\omega} := \lim_{N \to \infty} \frac{(\omega|_{C_N}) * \widetilde{(\omega|_{C_N})}}{(2N+1)^2}, \tag{2.3}$$

where C_N stands for the closed centred square of side length $2N$. Here, $\tilde{\mu}$ denotes the measure defined by $\tilde{\mu}(g) = \overline{\mu(\tilde{g})}$ for $g \in C_{\mathsf{c}}(\mathbb{R}^2)$, with $\tilde{g}(x) := \overline{g(-x)}$ (and where the bar denotes complex conjugation). The autocorrelation measure γ is of the form $\gamma = \eta\delta_{\mathbb{Z}^2}$ with autocorrelation coefficients

$$\eta(m,n) = \lim_{N \to \infty} \frac{1}{(2N+1)^2} \sum_{k,\ell=-N}^{N} w_{k,\ell} w_{k-m,\ell-n}. \tag{2.4}$$

All limits exists due to the unique ergodicity of the underlying dynamical system [6], under the action of the group $\mathbb{Z}^2$.

Clearly, one has $\eta(0,0) = 1$, while Eq. (2.1) implies the nine recursion relations

$$\begin{aligned}
\eta(3m,3n) &= \eta(m,n), \\
\eta(3m,3n+1) &= -\frac{2}{9}\eta(m,n) + \frac{1}{3}\eta(m,n+1), \\
\eta(3m,3n+2) &= \frac{1}{3}\eta(m,n) - \frac{2}{9}\eta(m,n+1), \\
\eta(3m+1,3n) &= -\frac{2}{9}\eta(m,n) + \frac{1}{3}\eta(m+1,n), \\
\eta(3m+1,3n+1) &= -\frac{2}{9}\big(\eta(m+1,n) + \eta(m,n+1)\big) + \frac{1}{9}\eta(m+1,n+1),
\end{aligned} \tag{2.5}$$

$$\begin{aligned}
\eta(3m+1,3n+2) &= -\frac{2}{9}\big(\eta(m,n)+\eta(m+1,n+1)\big)+\frac{1}{9}\eta(m+1,n),\\
\eta(3m+2,3n) &= \frac{1}{3}\eta(m,n)-\frac{2}{9}\eta(m+1,n),\\
\eta(3m+2,3n+1) &= -\frac{2}{9}\big(\eta(m,n)+\eta(m+1,n+1)\big)+\frac{1}{9}\eta(m,n+1),\\
\eta(3m+2,3n+2) &= \frac{1}{9}\eta(m,n)-\frac{2}{9}\big(\eta(m+1,n)+\eta(m,n+1)\big),
\end{aligned}$$

which hold for all $m,n\in\mathbb{Z}$ and determine all coefficients uniquely [6]. The autocorrelation coefficients show a number of remarkable properties, which are interesting in their own right, and useful for explicit calculations.

Since the support of ω is the lattice $\mathbb{Z}^2$, the diffraction measure $\hat{\gamma}$ is $\mathbb{Z}^2$-periodic [2], and can thus be written as

$$\hat{\gamma}=\mu*\delta_{\mathbb{Z}^2},$$

where μ is a positive measure on the fundamental domain $\mathbb{T}^2=[0,1)^2$ of $\mathbb{Z}^2$. One can now analyse $\hat{\gamma}$ via the measure μ, which, via the Herglotz–Bochner theorem, is related to the autocorrelation coefficients by Fourier transform

$$\eta(k)=\int_{\mathbb{T}^2} e^{2\pi\mathrm{i}kz}\,\mathrm{d}\mu(z),$$

where $k=(m,n)\in\mathbb{Z}^2$ and kz denotes the scalar product. We now sketch how to determine the spectral type of μ, and how to calculate it.

Defining $\Sigma(N):=\sum_{m,n=0}^{N-1}\eta(m,n)^2$, the recursions (2.5) lead to the estimate

$$\Sigma(3N)\le\frac{319}{81}\Sigma(N),$$

so that $\Sigma(N)/N^2\longrightarrow 0$ as $N\to\infty$. An application of Wiener's criterion in its multidimensional version [6, 7] implies that μ, and hence also the diffraction measure $\hat{\gamma}$, is continuous, which means that it comprises no Bragg peaks at all.

Since $\eta(0,1)=\eta(1,0)=-1/3$, which follows from Eq. (2.5) by a short calculation, the first recurrence relation implies that $\eta(0,3^j)=\eta(3^j,0)=-1/3$ for all integer $j\ge 0$. Consequently, the coefficients cannot vanish at infinity. Due to the linearity of the recursion relations, the Riemann–Lebesgue lemma implies [6] that μ cannot have an absolutely continuous component (relative to Lebesgue measure). The measure μ, and hence $\hat{\gamma}$ as well, must thus be purely singular continuous.

2.4 Riesz Product Representation

Although the determination of the spectral type of $\hat{\gamma}$ is based on an abstract argument, the recursion relations (2.5) hold the key to an explicit, iterative calculation of μ (and hence $\hat{\gamma}$). One defines the distribution function $F(x,y):=$