APERIODIC ORDER
Volume 2: Crystallography and Almost Periodicity

Quasicrystals are non-periodic solids that were discovered in 1982 by Dan Shechtman, Nobel Laureate in Chemistry 2011. The mathematics that underlies this discovery or was stimulated by it, which is known as the theory of Aperiodic Order, is the subject of this comprehensive multi-volume series.

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The volume covers various aspects of crystallography, generalising appropriately from the classical case to the setting of aperiodically ordered structures. A strong focus is placed upon almost periodicity, a central concept of crystallography that captures the coherent repetition of local motifs or patterns, and its close links to Fourier analysis, which is one of the main tools available to characterise such structures. The book opens with a foreword by Jeffrey C. Lagarias on the wider mathematical perspective and closes with an epilogue on the emergence of quasicrystals from the point of view of physical sciences, written by Peter Kramer, one of the founders of the field on the side of theoretical and mathematical physics.

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ENCYCLOPEDIA OF MATHEMATICS AND ITS APPLICATIONS

Aperiodic Order

Volume 2: Crystallography and Almost Periodicity

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Shaftesbury Road, Cambridge CB2 8EA, United Kingdom

One Liberty Plaza, 20th Floor, New York, NY 10006, USA

477 Williamstown Road, Port Melbourne, VIC 3207, Australia

314–321, 3rd Floor, Plot 3, Splendor Forum, Jasola District Centre, New Delhi – 110025, India

103 Penang Road, #05–06/07, Visioncrest Commercial, Singapore 238467

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www.cambridge.org
Information on this title: www.cambridge.org/9780521869928

DOI: 10.1017/9781139033862

First published 2017

A catalogue record for this publication is available from the British Library

ISBN 978-0-521-86992-8 Hardback

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Foreword by Jeffrey C. Lagarias

The mathematical study of aperiodically ordered structures is a beautiful synthesis of geometry, analysis, algebra and number theory. On the mathematical side, it arose in connection with tilings as a model of computation (the undecidability of the domino problem of Hao Wang) and the existence of ever simpler aperiodic tilings, exemplified by the Penrose tiling. From the physics side, it received great impetus from the discovery of Dan Schechtman in 1982 (published in 1984, Nobel Prize in Chemistry 2011) of an AlMn alloy whose X-ray diffraction spectrum exhibited long-range order of atomic positions and spacings with icosahedral symmetry.[1] That is, the sample exhibited an X-ray diffraction pattern with sharp spots with 10-fold, 6-fold and 2-fold symmetries when rotated to the corresponding directions of the icosahedron. Such a symmetry is incompatible with the material having an atomic structure that is periodic in any direction.

This discovery raised several questions, such as:

(1) Do ideal structures exist that have diffraction spectra with sharp spots and (perfect) non-crystallographic symmetries?

(2) Are there ‘local conditions’ permitting or favouring the assembly of such structures?

These two questions received positive answers in the 1980s, in the sense of mathematical constructions which achieve all or most of them. One such construction leads to cut and project sets and so-called model sets, which are described in detail in the first volume of this series [AO1]. There was earlier theoretical work anticipating these structures by various people, including Mackay (1981), Kramer (1982) as well as Kramer and Neri (1984).

There remain serious mathematical problems in extending these answers to a larger range of validity, including:

(i) Construction of interesting point sets;

(ii) Determination of local (matching) rules to force aperiodicity;

(iii) Classification of the possible types of symmetry.

[1] The corresponding references can be found in the bibliography of the first volume in this series [AO1]. Selected additional or new references will be given explicitly.

Answering these questions motivated the development of an extension of classical crystallography that is suitable to describe such structures, nowadays known as quasicrystals. In addition, establishing new notions of 'equivalence' of (aperiodic) structures requires new concepts. For instance, this task led to developments in ergodic theory with larger group actions, such as $\mathbb{Z}^d$, $\mathbb{R}^d$, or the Euclidean isometry group $\mathbb{R}^d \rtimes \mathrm{O}(d)$.

Besides the two questions above, there is a third question, concerning the inverse problem of reconstructing information on the atomic structure from diffraction data or from scanning tunneling electron microscope data. This amounts to asking: 'Where are the atoms?', which still seems a difficult problem to handle.

There are two major types of structures studied in aperiodic order. The first type consists of *Delone set models*, which concern uniformly discrete sets of points modelling the solid state, often imposing restrictions on allowable interpoint distance vectors. The second type consists of *tiling models*, where one studies tilings of Euclidean space with a finite number of distinct tile shapes, often polyhedra. In some of these models, additional 'matching rules' are imposed on how tiles may be placed next to each other. There are methods for taking a model structure of one type and converting it to the other type. For Delone sets, one may associate to it the tiling of space given by the Voronoi cells around its points. For a tiling model, one may mark a few points in the interior of each tile to assign a Delone set to the tiling.

Generalisation of geometric crystallography

The subject of *geometric crystallography* [5] was developed in the 19th century. Based on atomistic concepts, it considered infinitely extended discrete sets of points $\varLambda$ in space $\mathbb{R}^d$, called *regular point systems*, which are discrete sets that 'look the same' when centred at any point in them. That is, the set $\varLambda$ is preserved by any translation mapping one point of it to any other. A foundational result is that any such system of points must form a single (full-dimensional) lattice $\varGamma$ of points in $\mathbb{R}^d$. One may then classify such systems according to their full set of Euclidean symmetries (allowing reflections). This was accomplished for two dimensions in the 1870s and in three dimensions in independent work of Federov (1891), Schoenflies (1893) and Barlow (1893).

The problem of establishing a finite classification in d dimensions was raised by Hilbert in 1900 as part of his 18th problem. The first step to its solution was contributed by Bieberbach, who by 1912 showed that there are only finitely many symmetry types in each dimension. The classification in four dimensions was completed in 1978 by Brown, Bülow, Neubüser, Wondratschek and Zassenhaus.

A generalisation of regular point systems is that of *multiregular point systems*. These are infinitely extended discrete sets in $\mathbb{R}^d$ which, when centred at any point, are isometric to one of a finite list of such systems (fixing a marked centre point). Dolbilin et al. [3] showed that any such system necessarily is the union of a finite number of translates of a full-dimensional lattice $\varGamma$, so is fully periodic (or crystallographic).

To allow aperiodic point patterns, one enlarges the set of crystallographic point sets to Delone sets, or (r, R) sets, which are infinitely extended sets such that no two points are closer than distance r and such that each ball of radius R contains at least one point. There is a useful taxonomy on Delone sets that nicely extends the framework of geometric crystallography.

The first idea is to generalise the notion of regular point system to require the agreement of patches of a finite radius, rather than all the way to infinity. For a fixed set $\varLambda$, a patch of radius T centred at $x \in \varLambda$ is the set $\varLambda \cap (B_T(0) + x)$. In 1976, Delone and coworkers showed that regular point systems could be characterised by the property that they locally 'look the same' when centred around each point out to a sufficiently large finite radius T, where T is a function only of the Delone set parameters (r, R) and the dimension d. An extension of this result holds for multiregular point systems as well: If $\varLambda \subset \mathbb{R}^d$ is a Delone set that has exactly k different isometry classes of centred patches of a given radius T, with T sufficiently large with respect to k, namely $T \geqslant CRk$ with $C = 2(d^2+1)\log_2(2(R/r)+2)$, then $\varLambda$ is a multiregular point system having k different isometry classes of (infinite radius) patch types [3].

The second idea is to restrict the allowable interpoint distance vectors. The class of *finite local complexity* (FLC) Delone sets comprises those Delone sets $\varLambda$ for which $\varLambda - \varLambda$ is a discrete and closed set. In fact, it suffices to check this condition out to a finite radius $2R$: One only needs that $(\varLambda - \varLambda) \cap B_{2R}(0)$ is a finite set. One consequence is that the points of FLC Delone sets can be labelled by 'coordinates' in a finite-dimensional module, embedded in a space of dimension higher than that of the ambient space of the Delone set.

To go one step further, one considers *Meyer sets* in $\mathbb{R}^d$, which form the subclass of Delone sets $\varLambda$ for which the interpoint distance set (or Minkowski difference) $\varLambda - \varLambda$ is a Delone set. This version has been shown to be equivalent to Meyer's original notion [11], which is that of relatively dense sets $\varLambda$ such that $\varLambda - \varLambda \subseteq \varLambda + F$ with F a finite set.

A quantification of order for FLC Delone sets is provided by numerical combinatorial invariants of the structure of their finite patches. The *patch-counting function* $f(R)$ counts the number of translation-inequivalent patches of radius R. The growth rate of this function with increasing R is a combinatorial measure of the possible kind of order. It has been shown by Lagarias and Pleasants (2002) that, if this growth rate is sublinear, the function must be eventually constant and the structure must then be an ideal crystal. A second quantifier concerns measures of distances for the repetition of different patches of large radius in the set. One says that an FLC Delone set $\varLambda$ is *linearly repetitive* if there is a constant C such that every fixed patch of radius ρ that occurs somewhere in the set necessarily occurs within distance $C\rho$ of any point of $\varLambda$. This condition holds for Penrose tilings, for example, and it also implies that such a set must have a well-defined diffraction measure.

Inflation rules

A frequently used construction for aperiodic patterns employs structures that are (possibly approximately) preserved under an inflation operation. Meyer's work in harmonic analysis from the early 1970s included a study of discrete systems which may, in special cases, be preserved under an inflation rule, which reproduces a structure on a larger scale. He noted a connection between allowable inflation scales on these structures and algebraic numbers.

A point set $\varLambda$ has an *inflation* if there is a number $\eta > 1$ such that $\eta\varLambda \subset \varLambda$; we call any such η an *inflation factor* for $\varLambda$. Meyer [12] proved for the sets which are now called Meyer sets that the inflation factor must be an algebraic integer which is either a Pisot–Vijayaraghavan (PV) number (all algebraic conjugates satisfy $|\eta'| < 1$) or a Salem number (all $|\eta'| \leqslant 1$ and some $|\eta'| = 1$). The golden ratio $\tau = \frac{1+\sqrt{5}}{2}$ is a PV number. It features in the mathematics of the icosahedron and appears in all tiling models with icosahedral symmetry as well as in fivefold symmetric tilings of the plane. Later, in 1999, I observed that there is also an algebraic restriction on inflation factors $\eta > 1$ of FLC Delone sets: They must be real algebraic integers all of whose algebraic conjugates η' satisfy $|\eta'| \leqslant \eta$.

Packing problems and quasicrystallinity

Packing problems have been observed to possess connections with crystallography. The general packing problem includes the determination of the densest packings attainable by identical copies of fixed solid geometric objects, particularly convex bodies. Minkowski's 'Geometry of Numbers' concerns the problem of finding the densest lattice packing of identical copies of a given convex body, movable by translations only. Allowing rotations of the body, as in the case of tetrahedra, leads to new problems.

Notable examples have densest packings attainable by a crystalline structure. For equal spheres, the densest packing in dimensions up to three are all attained by lattice packings. In three dimensions, there are also equally dense periodic packings of various types, as well as packings that are aperiodically stacked in one direction while being periodic in the two independent directions orthogonal to it.

Recent developments suggest that suitable tiling questions may also lead to quasicrystalline structures. For packings of regular tetrahedra, there are no mathematical proofs but there are results obtained by simulation. The densest known packing of regular tetrahedra has a periodic structure with four tetrahedra in the unit cell [2]. On the other hand, Monte Carlo simulations of Haj-Akbari and coworkers [6] of a 'gas' of regular tetrahedra at high pressure (meaning ensembles having density close to this maximal value) suggest they have a quasicrystalline structure in two directions, while having a periodic structure in the third direction. Specifically, samples displayed a diffraction pattern (for point scatterers located at centroids of the tetrahedra) that exhibits a ring of peaks indicating a 12-fold symmetry.

Diffractivity

The study of diffractivity properties of aperiodic sets requires Fourier analysis and distribution theory. Here, we only consider diffraction for point sets in $\mathbb{R}^d$, although Meyer — and later Moody as well as Schlottmann — have shown that the analysis of diffraction can profitably be done in the more general setting of locally compact Abelian groups. The formulation of a general mathematical notion of diffractivity suitable for diffraction of aperiodic sets (via a connection with ergodic theory) was initiated by Dworkin (1993) and extended by Hof (1995). It uses a framework of locally finite measures, which for $\mathbb{R}^d$ can be viewed as a subclass of tempered distributions. An autocorrelation measure is associated to a given spatial distribution of δ-functions as a locally finite measure; see [AO1, Chs. 8 and 9] for a detailed exposition. The diffraction data is the Fourier transform of this measure, viewed as a positive definite measure. A set will be called *pure point diffractive* if this Fourier transform is itself a pure point measure, where the spectrum may be a dense set of points.

A special case of this notion of diffractivity is given by the *Poisson summation formula* (PSF). Given a lattice $\varGamma \subset \mathbb{R}^d$, consider the locally finite measure (Dirac comb) $\delta_{\varGamma} := \sum_{x\in\varGamma} \delta_x$, where δ_x is the normalised Dirac measure at x. In this case, the autocorrelation measure of $\delta_{\varGamma}$ is a scaled multiple of $\delta_{\varGamma}$. Its Fourier transform[2] is a (different) scaled multiple of $\delta_{\varGamma^*}$, where $\varGamma^*$ is the dual lattice of $\varGamma$ A generalisation of this formula shows that

[2]The PSF is more commonly written as the evaluation of a function against this tempered distribution $\delta_{\varGamma}$, in which one side of the formula is the sum $\sum_{x\in\varGamma} f(x)$ and the other side is a weighted sum of the Fourier transform of f evaluated on the dual lattice.

an ideal crystal is pure point diffractive, with its spectrum supported on a Delone set, the dual lattice $\varGamma^*$.

The cut and project construction leads to many Meyer sets with pure point diffraction spectrum. In particular, regular model sets have this property. A formula for the diffraction of cut and project sets was independently found by many people, including Elser (1986). A mathematically rigorous approach was developed later, starting with work by de Bruijn (1986), Hof (1995) and Schlottmann (2000). The Delone set condition on the initial set can be relaxed, as demonstrated for the visible lattice points in $\mathbb{R}^d$ by Baake, Moody and Pleasants (2000). This set is pure point diffractive, but not relatively dense.

My survey [7] from 2000 discussed results on diffractivity and their relation to classes of almost periodic functions. That paper formulated questions concerning the existence of pure point measures supported on Delone sets having pure point diffraction measures with uniformly discrete or Delone set support. This research area is active and has had substantial recent advances. In 2015, Lev and Olevskii [9] showed that, in the one-dimensional case, all such measures come from the Poisson summation formula. For higher dimensions, there are many exotic examples, found by Favorov, Lev and Olevskii, Meyer, and Kolountzakis; see [13] and references therein. Further advances in both directions are made in [10].

The diffraction spectrum of various aperiodic sets which possess an inflation factor has been much studied. Some of these sets have pure point spectrum, in other cases they have mixed spectrum. It is an open problem whether such inflation sets, when their autocorrelation is a pure point measure, must necessarily have an inflation factor that is a PV number. In another direction, the diffraction spectrum of a Delone set can be related to to the dynamical spectrum of an associated dynamical system with a translation action by $\mathbb{R}^d$ (or, in the lattice-periodic setting, by $\mathbb{Z}^d$); see [1] for a survey.

A famous inverse problem for X-ray diffraction is that of reconstructing the atomic structure of a periodic crystal from X-ray diffraction data. This problem requires overcoming the difficulty that diffraction data determine the intensities of spots but lose the phase information.[3] Consequently, the diffraction image cannot tell certain periodic structures apart. Such structures are called *homometric* and were studied by Pauling and Patterson in the 1930/40s. A Nobel Prize in Chemistry was awarded in 1985 to Hauptmann and Karle ‘for outstanding achievements in the development of direct

[3] In terms of Fourier transforms, the scattering intensities record the squared absolute values of the Fourier amplitudes (or coefficients).

methods in the determination of crystal structure' to recover phase information. It is an open problem to determine suitable 'phase information' that might be associated with a diffractive aperiodic set; see [8] for first steps towards a classification. In this context, it is also important to investigate how modulated structures can be distinguished. These questions suggest the investigation of new classes of almost periodic functions.

Connections with number theory

There is an unreasonably effective connection of quasicrystalline structures with algebraic number theory, which already appears in the title of Meyer's 1972 book [11] in which he introduced the notion of what one now calls model sets (simply called 'model' there).

Classical problems in number theory produce crystalline structures with extra symmetries given by Galois group actions. Consider the ring of integers $\mathcal{O}_K$ of an algebraic number field K for which K is a Galois extension of the rational numbers $\mathbb{Q}$ with (finite) Galois group G. Such a ring of integers possesses a Minkowski embedding, compare [AO1, Sec. 3.4], as a lattice in a suitable Euclidean space $\mathbb{R}^d$ (of dimension $d = [K : \mathbb{Q}]$), in such a way that the symmetries of the Galois group G act linearly on the coordinates of this Euclidean space, and leave the lattice $\mathcal{O}_K$ invariant. For example, take G to be the alternating group A_5 of order 60, which is the rotation symmetry group of the regular icosahedron and the smallest non-Abelian simple group. One can find an irreducible equation of degree 5 over $\mathbb{Q}$ whose splitting field (normal Galois closure) has group A_5. The ring of integers of the normal closure of this field then carries an action of A_5, and the restriction to a suitable sublattice can give an (inefficient) cut and project construction.

This lattice embedding of algebraic integers was used by Minkowski in his 'Geometry of Numbers'. His study of lattice packings of convex bodies was invented, in part, to prove results in number theory related to bounds for discriminants of number fields and finiteness of class numbers of algebraic number fields.

The Poisson summation formula plays an important role in number theory, connecting it to harmonic analysis. The functional equation of the Riemann zeta function encodes the PSF in one dimension, and vice versa. The property of pure point diffractivity for certain lattice Dirac combs is another instantiation of the PSF. The existence of quasicrystals which appear to have pure point diffraction spectrum hints at the existence of new kinds of summation formulas generalising the PSF, a problem raised by Dyson [4], who asked whether it might shed light on the Riemann hypothesis. The 'explicit formulas' of prime number theory have a form resembling the PSF, preserving discreteness of point sets but not preserving the Delone set property.

Many of the topics above have been introduced and discussed in [AO1]. The present volume presents chapters surveying and extending several of these topics. The first chapter studies inflation tilings, and the second chapter considers the problem of reconstructing the parameters of model sets from tomographic data. The subsequent chapter considers enumeration problems for embedded sublattices which are related to crystallographic questions. Three further chapters present a detailed account of the structure of almost periodic measures, in a form useful for advancing the study of diffractivity of aperiodic structures. The volume concludes with an epilogue on the physical precursors to the discovery of quasicrystals.

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Preface

This is the second volume in a series of books exploring the mathematics of aperiodic order. While the first volume was meant as a general introduction to the field, we now start to develop the theory in more depth. To do justice to the rapidly expanding field, we decided to work with various authors or teams of authors, which means that this book is somewhere intermediate between a monograph and a review selection. Future volumes will also be structured in this way.

Clearly, almost periodicity is a central concept of crystallography, as it reflects and captures the coherent repetition of local motifs or patterns. The foremost tool to analyse such structures is provided by Fourier analysis of measures, which thus forms a substantial part of this volume. Other important aspects are usually analysed by group theoretic or general algebraic methods. In this respect, due to the availability of comprehensive reviews and several books, we decided to not include a chapter on space groups and their generalisation to quasicrystals.

The main text begins with a chapter on inflation tilings, contributed by Dirk Frettlöh. It augments the discussion of the first volume by presenting a panorama of less familiar constructions and recent developments. This is followed by a contribution to the inverse problem of discrete tomography, where special emphasis lies on the comparison between notions from classical (periodic) crystallography and their extensions to quasicrystals. A similar interplay is prevalent in the ensuing chapter on enumeration problems for lattices versus embedded $\mathbb{Z}$-modules, which highlights the power of number-theoretic methods in the theory of aperiodic order.

The substantial part on almost periodicity and its facets begins with a thorough exposition of the general theory of almost periodic measures on locally compact Abelian groups, contributed by Robert V. Moody and Nicolae Strungaru. This comprehensive summary emerged from the need to understand the spectral structure of aperiodic systems. Perhaps the most important connection exists with the structure of Meyer sets and their description via cut and project schemes, which is developed in the ensuing chapter by Nicolae Strungaru. This part is concluded by an expository discussion of the sampling problem for (almost) periodic functions along exponential sequences, which highlights yet another connection with number theory.

Complementing the foreword by Jeffrey C. Lagarias, this volume ends with an epilogue on the emergence of quasicrystals from the perspective of physical sciences, with a focus on the underlying theoretical ideas. This epilogue was contributed by Peter Kramer who is one of the pioneers and founders of the field on the side of theoretical and mathematical physics.

As mentioned above, this volume consists of solicited reviews and thematic additions. All chapters have been edited or partly redrafted by us to match the style of the series and its general notation as far as possible. We have thus made the first volume in the aperiodic order series the main reference for all chapters, and refer to it frequently. Nevertheless, some deviation and/or additions are inevitable as a consequence of the established conventions in different mathematical disciplines. Some other, more minor changes have also occurred, such as distinguishing between inclusion and proper inclusion of sets. Where appropriate, such modifications are detailed in footnotes. Each chapter has its own bibliography, while the general index covers all chapters and is also meant to reflect connections between the expositions.

Let us give some background on the tiling that is shown on the book cover. It was designed by Franz Gähler, and is locally equivalent (in the sense of mutual local derivability) to his shield tiling. It was originally designed for a competition at the Fields Institute for Research in Mathematical Sciences in Toronto in 1995. In this year, the Fields Institute moved from Waterloo to Toronto, where a fundraising tiling on the wall in the backyard was planned. Gähler's submission won the competition, but, for a number of reasons, the tiling wall was never realised.

Various people have favourably contributed to this volume. First of all, we would like to thank all authors for the effort they have put into the individual chapters, and into critically reading and commenting on other parts of the volume. Furthermore, we are indebted to Franz Gähler, Neil Mañibo, Yasushi Nagai, Dan Rust, Timo Spindeler, Venta Terauds and Christopher Voll for their comments and suggestions, which helped to improve the exposition. Special thanks also to Franz Gähler for providing the cover illustration. Last but not least, we thank the staff from Cambridge University Press for an always smooth cooperation, the German Research Council (DFG) for support through CRC 701, and the School of Physical Sciences at the University of Tasmania in Hobart for its hospitality during several visits, which helped us immensely to complete this volume.

Michael Baake and Uwe Grimm