



Quasicrystals

Fundamentals and Applications

Enrique Maciá-Barber



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Preface

During the early 1980s, a few people started to realize that certain arrays of atoms *lacking* periodic translation symmetry were surprisingly able to give rise to sharp diffraction patterns, fully constellated with a lot of discrete Bragg reflections, just as well-known periodic arrays of atoms generally do. In addition, their diffraction patterns revealed a *new crystallographic symmetry*, namely, scaling invariance, which endows them with an impressive esthetical appeal. This symmetry had not been previously considered in classical crystallography, and it is directly related to the emergence of self-similar, hierarchical patterns embodying atomic cluster aggregates. Therefore, from a structural viewpoint these compounds can be regarded as self-similar arrays of atoms, where the translation symmetry, characteristic of periodic crystals, is replaced by a scale invariance one. In this sense, they could be pictured as fractal solids representatives, with scale factors given by *irrational* numbers. Consequently, this novel condensed matter phase can be properly regarded as describing long-range ordered solids whose diffraction patterns exhibit symmetries that are incompatible with translational symmetry.

Upon closer scrutiny it was understood that these solids represent a natural extension of the periodic crystal notion when the location of atoms throughout the space is described in terms of *quasiperiodic functions* instead of the usual periodic ones. Accordingly, they were referred to as quasiperiodic crystals – *quasicrystals*, for short. In retrospective, one may consider the choice of this abbreviation as somewhat unfortunate. In fact, the very semantics of the term *quasicrystal*, albeit originally derived from the mathematically rigorous quasiperiodic function notion, suggested that quasicrystals (QCs) were occupying an intermediate position between well ordered arrangements of matter and glassy state ones, thereby denoting some sort of defective, flawed materials which failed to properly fulfil the basic tenets of well behaved condensed matter representatives. Quite on the contrary, it is now well established that QCs show up long-range orderings of atoms rendering essentially discrete diffraction patterns, which can be indexed in terms of a hyperspace generalization of classical crystallography, suitably including inflation symmetry operations. Therefore, the discovery of QCs ultimately led to the convenience of revamping the very notion of crystal in condensed matter.

Quasicrystals also exhibit unusual physical properties, closely related to the fractal nature of their energy and frequency spectra. Physically this feature means that some specific fragments of the spectra appear once and again at different scales. Thus, rather than trying to fit the electronic structure of QCs within the conceptual schemes introduced in order to describe classical periodic solids (namely, metals, semimetals, semiconductors, or insulators) we should expand these categories to properly deal with their highly-fragmented energy and frequency spectra. Indeed, it has been confirmed that transport properties of thermodynamically stable QCs of high structural quality resemble a more semiconductor-like than metallic character. Therefore, QCs provide an intriguing example of ordered solids made of typical metallic atoms which do not exhibit most of the physical properties usually signaling the presence of metallic bonding. Accordingly, by the light of the knowledge gained during the years elapsed, it is now clear that QCs are representatives of a novel phase of matter exhibiting a large number of remarkable specific properties.

This book aims at providing a detailed and updated introduction to the field of QCs in an interdisciplinary way, focusing on their fundamental aspects but also on their present and future applications. The role of quasiperiodic order in science and technology is also examined by highlighting the new design capabilities provided by this novel ordering of matter. This book is specifically devoted to promoting the very notion of quasiperiodic order in condensed matter, and to spur its physical implications and technological capabilities in materials science. It, therefore, explores the main topics related to intermetallic, photonic and phononic quasicrystals, as well as soft-matter quasicrystals, by the light of their intrinsic physical and structural properties. It also explores exciting applications of quasiperiodically ordered systems in new technological devices, including multilayered quasiperiodic systems, along with 2D and 3D designs. To this end, I will thoroughly discuss both experimental data and the most promising theoretical approaches aimed to explain them, properly comparing the main distinctive features of classical crystals and QCs from an analogical perspective.

Several topics on the role of aperiodic order in different domains of physical sciences and technology will be covered in this book. The first chapter address some basic notions and present the most characteristic features of different kinds of aperiodic systems in a descriptive way. In Chapter 1, we introduce different orderings of matter and describe the progressive transition from periodic to aperiodic thinking in physical sciences. In Chapter 2, the very notion of aperiodic crystal is introduced, fully describing its historical roots as well as the paramount discovery of quasicrystalline alloys and their beautiful novel symmetries. The study of the unusual physical properties of quasicrystalline alloys is then presented in more detail in Chapter 3, paying a special attention to their intriguing electronic structure and the possible nature of chemical bonding in hierarchically arranged cluster based solids. In Chapter 4, we introduce the basic structural properties of man-made materials consisting of aperiodic sequences of layers such as Fibonacci semiconductor-based superlattices or Cantor-like dielectric multilayers. The main mathematical features of the substitution sequences defining their growth rule are also reviewed along with the possible signatures of quasiperiodicity in their physical properties. In Chapter 5, we focus on the possible applications of QCs, and in Chapter 6, we outlook new frontiers in the QCs science in the years to come.

The book contains 50 proposed exercises (highlighted in boldface through the text) accompanied by their detailed solutions, motivating and illustrating the different concepts and notions to provide readers with further learning opportunities. I have prepared the exercises mainly from results published and discussed in regular research papers during the last decade, in order to provide a glimpse into the main current trends in the field. Although the exercises and their solutions are given at the end of each chapter for convenience, it must be understood that they are an integral part of the presentation. Accordingly, it is highly recommended to the reader that he/she try to solve the exercises in the sequence they appear in the text, then check his/her obtained result with those provided at the end of the chapter, and only then to resume the reading of the main text. In this way, the readers (who are intended to be both graduate students as well as senior scientists approaching this rapidly growing topic from other research fields) will be able to extract the maximum benefit from the materials contained in this book in the shortest time. All the references are listed in the bibliography section at the end of the book. I have tried to avoid a heavily referenced main text by concentrating most references in the places where they are most convenient to properly credit results published in the literature, namely, in the figures and tables captions, in the footnotes, and in the exercises and their solutions.

This book is dedicated to the memory of Professors Esther Belin-Ferré, Patricia A. Thiel and An Pang Tsai, with my heartfelt gratitude for the continued interest in my research activities, valuable advice and motivating support they provided me during the time I had the fortune to share with them.

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Enrique Maciá Barber
Madrid, July 2020

Intermetallic Quasicrystals

1.1 WHAT IS A QUASICRYSTAL?

“A quasicrystal is the natural extension of the notion of a crystal to structures with quasiperiodic, rather than periodic, translational order”. (Dov Levine and Paul Joseph Steinhardt, 1984) [486]

1.1.1 The $\text{Al}_{86}\text{Mn}_{14}$ alloy electron diffraction picture

“It took the right instrument, an electron microscope, because while X-ray diffraction is great for studying many aspects of crystals, it cannot show you rotational symmetry” (Daniel Shechtman, 70th Birthday Celebration Symposium, 27 July 2011, Iowa State University, Ames).

“As I was studying rapidly solidified aluminium alloy which contained 25% manganese by transmission electron microscopy, something very strange and unexpected happened. There were 10 bright spots in the selected area diffraction pattern, equally spaced from the center and from one another. I counted them and repeat the count in the other direction and said myself: ‘There is no such animal’. In Hebrew: ‘Ein Chaya Kazo’”. (Daniel Shechtman commenting on his 8 April 1982 discovery [321], see Fig. 1.1)

To properly understand Daniel Shechtman’s astonishment, we must recall that 20th century crystallography started in 1912 with the introduction of X-ray diffraction methods by Max von Laue (1879–1950), William Henry Bragg (1862–1942), and William Lawrence Bragg (1890–1971), who assumed that atoms were arranged in ordered crystals according to regular *periodic* arrays on a lattice. During the seventy years elapsed until Shechtman’s discovery almost all reported crystalline structures were perfectly compatible with that assumption, and accordingly the main attribute of crystalline solid state was progressively identified with the presence of a periodic distribution of matter density, $\rho(\mathbf{r})$, inside the considered samples. Ideally assuming that atoms can be treated as material points, such a distribution can be envisioned as a lattice of points in Euclidean three-dimensional space, so that the vectors describing the lattice points have the general form $\mathbf{r} = n_1\mathbf{e}_1 + n_2\mathbf{e}_2 + n_3\mathbf{e}_3$, where $\{\mathbf{e}_i\}$ is a suitable vector basis and the coordinates are integer numbers (i.e., $n_i \in \mathbb{Z}$). Within this framework, lattice periodicity is given by the invariance condition $\rho(\mathbf{r} + \mathbf{R}_0) = \rho(\mathbf{r})$, where $\mathbf{R}_0 = m_1\mathbf{e}_1 + m_2\mathbf{e}_2 + m_3\mathbf{e}_3$ ($m_i \in \mathbb{Z}$) belongs to the lattice-related vectorial space. Physically, lattice periodicity guarantees that the local atomic environment we have at a given volume determined by the vector basis $\{\mathbf{e}_i\}$ inside the crystal exactly repeats itself throughout the space.

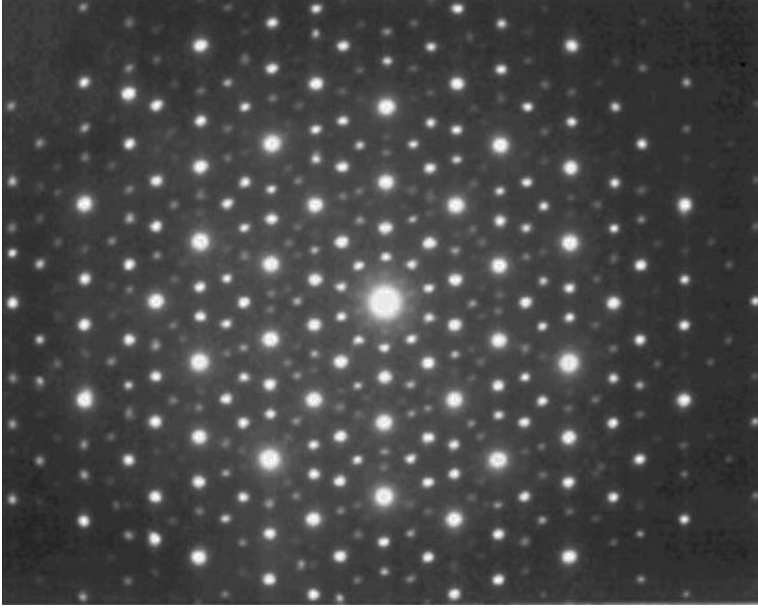


Figure 1.1 Electron diffraction pattern corresponding to the $\text{Al}_{86}\text{Mn}_{14}$ alloy quasicrystal discovered by Shechtman. A 10-fold symmetry axis around the origin can be clearly appreciated. (Reprinted figure with permission from Shechtman D, Blech I, Gratias D, and Cahn J W, *Physical Review Letters* 53, 1951, 1984. Copyright (1984) by the American Physical Society).

Periodic arrangements in Euclidean space involve rotations along with translations. We say that a given structure possesses a m -fold rotational symmetry if it is left unchanged when rotated by an angle $2\pi/m$, with respect to a certain axis, fixed a given point. The integer $m \in \mathbb{N}$ is called the order of the rotational symmetry (or of its related symmetry axis). Therefore, the lattice remains the same under rotations relating lattice points to each other, which are represented by a set of matrices of the form

$$\mathbf{R}_\alpha = \begin{pmatrix} n_{11} & n_{12} & n_{13} \\ n_{21} & n_{22} & n_{23} \\ n_{31} & n_{32} & n_{33} \end{pmatrix},$$

whose entries are integer numbers. On the other hand, on a properly chosen basis, pure rotations can be expressed in terms of the orthogonal matrices¹

$$\mathbf{M}_X = \begin{pmatrix} 1 & 0 & 0 \\ 0 & c & -s \\ 0 & s & c \end{pmatrix}, \quad \mathbf{M}_Y = \begin{pmatrix} c & 0 & s \\ 0 & 1 & 0 \\ -s & 0 & c \end{pmatrix}, \quad \mathbf{M}_Z = \begin{pmatrix} c & -s & 0 \\ s & c & 0 \\ 0 & 0 & 1 \end{pmatrix}, \quad (1.1)$$

with $c \equiv \cos \varphi$ and $s \equiv \sin \varphi$, describing counterclock rotations by an angle φ around the X , Y , and Z Cartesian axes, respectively. Accordingly, $\mathbf{R}_\alpha = \mathbf{B}\mathbf{M}_\alpha\mathbf{B}^{-1}$, where $\mathbf{B}$ denotes the basis matrix. Now, since the trace of a matrix is invariant under a basis transformation, the periodicity condition $\rho(\mathbf{R}_\alpha\mathbf{r}) = \rho(\mathbf{r})$ relating lattice points, implies that $\text{tr}(\mathbf{R}_\alpha) = \text{tr}(\mathbf{M}_\alpha)$,

¹Note that these matrices are unimodular (i.e., $\det \mathbf{M}_\alpha = 1$) as well.

TABLE 1.1 Allowed symmetry axes in 3D periodic crystals

n	φ	AXIS
-1	π	2-fold
0	$2\pi/3$	3-fold
1	$\pi/2$	4-fold
2	$\pi/3$	6-fold
3	0	Identity

so that we obtain the so-called crystallographical restriction [381, 840],

$$1 + 2 \cos \varphi = n_{11} + n_{22} + n_{33} \equiv n \in \mathbb{Z}, \quad (1.2)$$

which can be rewritten in the convenient form $\cos \varphi = (n - 1)/2$. The bound $|\cos \varphi| \leq 1$ then implies $n = \{-1, 0, 1, 2, 3\}$. In this way, we obtain the allowed rotation angles listed in Table 1.1. Therefore, a very small number of rotations are compatible with the periodicity condition, namely, only 2-, 3-, 4-, and 6-fold symmetry axes are allowed for 3D periodic lattices. Not surprisingly, Eq. (1.2) along with its corresponding version for two-dimensional (2D) periodic lattices (**Exercise 1.1**), have played a significant role in the foundations of classical crystallography. Accordingly, solid state physics textbooks properly emphasized that all rotational symmetries other than those listed in Table 1.1 were forbidden in crystals. On the basis of these allowed rotational symmetry elements, the so-called 14 Bravais lattices naturally followed, along with the related 230 space groups, thus providing the basic tool for crystal classification and the *International Tables of Crystallography* was the ultimate classification catalog for crystals. In this way, for many decades, it was firmly believed that 5-fold rotational symmetry could not exist in well ordered condensed phases of matter, so that the pentagon symmetry, which is widely found in the world of the living, was excluded from the mineral kingdom,² ... until Shechtman took his now celebrated electron diffraction picture shown in Fig. 1.1.³

Quite remarkably, the electron diffraction study of the $\text{Al}_{86}\text{Mn}_{14}$ alloy performed by Shechtman not only revealed the presence of this unexpected symmetry axis. In fact, when the specimen was tilted and viewed from other directions, the characteristic fingerprints of 2- and 3-fold symmetry axes were also observed. The complete account of symmetry rotation axes yield fifteen 2-fold axes, ten 3-fold axes, and six 5-fold axes, which together with the corresponding mirror planes, amount to the 120 symmetry elements characteristic of the *icosahedral* group, as it is shown in Fig. 1.2.

The existence of a novel solid phase of matter, which diffracts electrons like a single crystal does but has icosahedral point group symmetry, which is inconsistent with periodic lattice translations, was announced to the scientific community by Daniel Shechtman, Ilan Blech, Denis Gratias, and John W. Cahn (1928–2016) in a paper entitled “*Metallic phase with long-range orientational order and no translational symmetry*”, published in the *Physical Review Letters* on 12 November 1984 [801]. The remarkable sharpness of the diffraction patterns shown in Figs. 1.1 and 1.2 clearly indicates a high coherency of the electrons spatial

²Some observations of minerals probably exhibiting quinary axes of symmetry were earlier reported by Rome d’Isle, but by all indications these early observations were subsequently ignored [700].

³Because diffraction patterns are always inversion symmetric, it is convenient to distinguish between *diffraction* symmetry axes (10-fold in this case) and *real space* rotational symmetry axes (5-fold in this case). See Secs. 6.1.3.2 and 6.3.3.

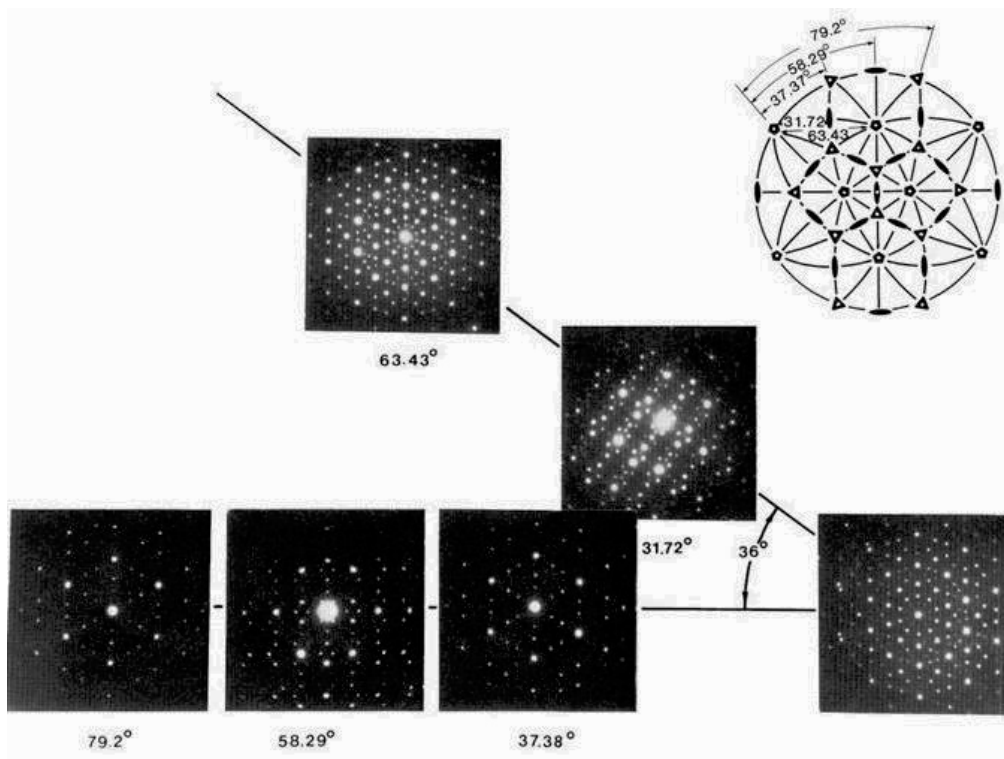


Figure 1.2 Selected-area electron diffraction patterns taken from a single grain of the $\text{Al}_{86}\text{Mn}_{14}$ alloy phase discovered by Shechtman. Rotations match those indicated in the stereographic projection of the symmetry elements of the icosahedral group $m\bar{3}5$ shown in the upper right corner. (Reprinted figure with permission from Shechtman D, Blech I, Gratias D, and Cahn J W, *Physical Review Letters* 53, 1951, 1984, Copyright (1984) by the American Physical Society).

interference, comparable to the one usually encountered in classical periodic crystals. Thus, this alloy exhibited well-defined long-range order, but one which is explicitly incompatible with the periodic translational one. What may be the nature of this ordering of matter which had remained so elusive to crystallographers until then?

Six weeks after Shechtman and co-workers paper publication, the nature of this long-range order was explained by Paul J. Steinhardt and Dov Levine in a paper entitled “*Quasicrystals: a new class of ordered structures*”, published in the *Physical Review Letters* on 24 December 1984 [486]. In this paper the notion of quasiperiodic crystals was introduced (see the quotation opening this Chapter) by invoking the mathematical notion of quasiperiodic functions (which will be described in Sec. 1.3.2), thereby widening the concept of a periodic distribution of atoms through the space to that of a quasiperiodic (QP) one.

This extension was performed on the basis of a detailed comparison between a computed diffraction pattern for a suitable QP atomic model and the electron diffraction pattern obtained by Shechtman for the $\text{Al}_{86}\text{Mn}_{14}$ alloy (Fig. 1.3). By inspecting both figures, it is apparent that the actual atomic arrangements in the alloy must be closely related to those adopted by the lattice points in the model. Indeed, the position of each electron diffraction reflection matches with the position of a peak in the calculated pattern, and a hierarchy of

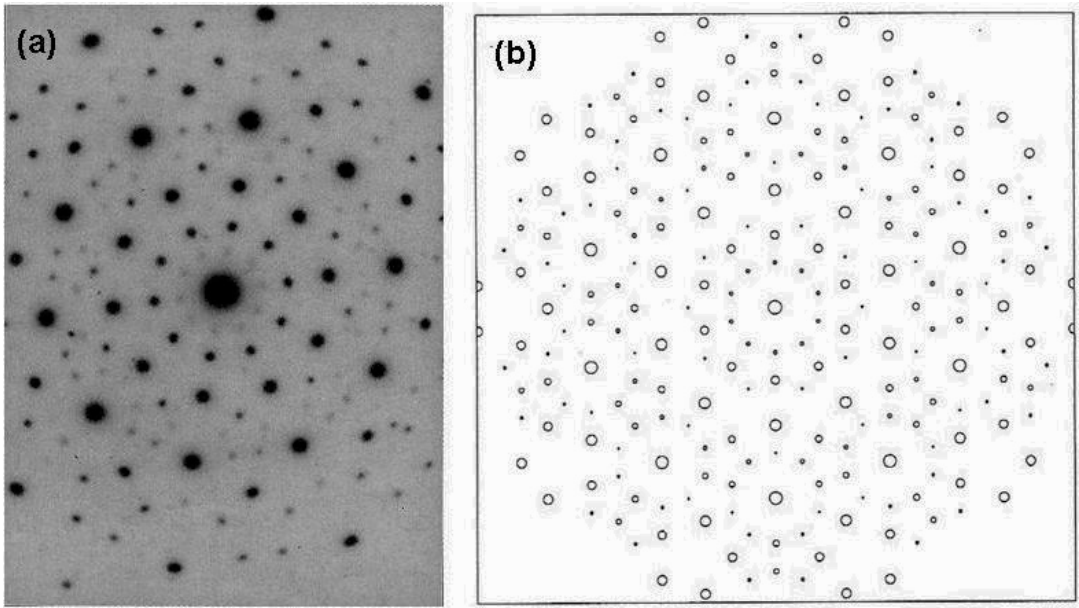


Figure 1.3 (a) Electron diffraction pattern corresponding to the $\text{Al}_{86}\text{Mn}_{14}$ QC discovered by Shechtman. (Reprinted figure with permission from Shechtman D, Blech I, Gratias D, and Cahn J W, *Physical Review Letters* 53, 1951, 1984. Copyright (1984) by the American Physical Society). (b) Computed diffraction pattern in a plane normal to a 5-fold axis for an ideal icosahedral QC, displaying only peaks above some given intensity. The circles are centered at the location of Bragg peaks and have a radius proportional to their intensity. (Reprinted figure with permission from Levine D and Steinhardt P J, *Physical Review Letters* 53, 2477, 1984. Copyright (1984) by the American Physical Society).

intensities characteristic of quasiperiodicity is observed at several orders [486]. In this way, the discovered novel phase was envisioned as a natural extension of the classical crystal notion, now embodying QP arrangements of matter as well. Accordingly, the shorthand *quasicrystal* (QC), standing for *quasiperiodic crystal*, was coined.⁴

Paul J. Steinhardt and Dov Levine were awarded the Oliver Buckley Condensed Matter Prize from the American Physical Society, in 2010 for “*pioneering contributions to the theory of quasicrystals, including the prediction of their diffraction pattern*”. Daniel Shechtman was awarded the Nobel Prize in Chemistry, in 2011 for the “*discovery of quasicrystals*”, almost thirty years after this discovery took place.

⁴The abstract of a paper submitted by David Ruelle on 13 January 1981 reads “*We discuss the possibility that, besides periodic and quasiperiodic crystals, there exist turbulent crystals as thermodynamic equilibrium states at non-zero temperature*” [760]. These quasiperiodic crystals exhibit “*long-range order with two independent modulations*” [760], thereby belonging to the family of incommensurate structures first described by Pim de Wolff and co-workers in Ref. [172]. Certainly, it is quite remarkable that D. Ruelle considers “*an ordinary crystal (periodic or quasiperiodic)*” as early as in 1981, hence anticipating the unifying view given in the revamped definition of crystal proposed by the ICrU in 1992 [357] (see Sec. 1.1.5).

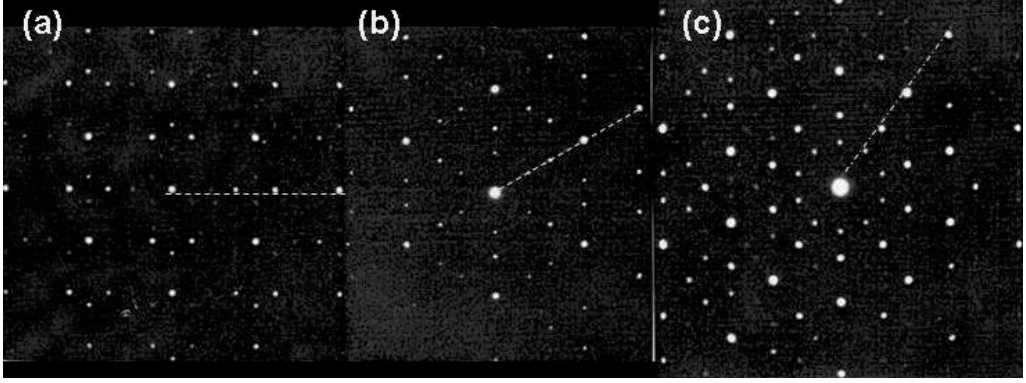


Figure 1.4 Electron diffraction patterns taken from an $i\text{-Al}_{86}\text{Mn}_{14}$ QC grain showing the 2-fold (a), 3-fold (b) and 5-fold (c) rotation axes of the icosahedral $m\bar{3}5$ point group. (Reprinted figure with permission from Elser V, Physical Review B 32, 4892, 1985. Copyright (1985) by the American Physical Society).

1.1.2 A novel crystallographic symmetry

“The diffraction pattern of icosahedral quasicrystals revealed a new crystallographic symmetry: scaling invariance”. (Aloysio Janner, 2006) [374]

In order to gain a deeper understanding on the nature of the spatial order underlying atomic arrangements in QCs, let us consider the electron diffraction patterns shown in Figs. 1.4 and 1.5. By inspecting the distribution of reflection peaks in these diffraction patterns one can observe the presence of:

- The full set of icosahedral point group symmetry elements, namely 2-, 3-, and 5-fold axes, along with the corresponding mirror planes, characteristic of the so-called *icosahedral* QCs (iQCs).
- The fingerprints of scale invariance symmetry, which can be determined from the study of two main features, namely, (1) a series of linear arrays of diffraction spots along the radial directions from the center peak (dashed lines in Fig. 1.4 and solid radial lines in Fig. 1.5), and (2) the presence of nested polygonal Bragg reflection patterns, featuring the so-called *Pythagorean pentagram* design (Fig. 1.5).

In order to disclose this characteristic inflation symmetry, let us start by closely inspecting Fig. 1.5. To this end, we measure the distances of successive main diffraction spots along a radial direction outwards from the center, d_n , and calculate the ratios d_n/d_1 . The d_n obtained values are listed in the second column of Table 1.2, and they clearly follow a non-periodic series, which obeys the recursion rule $d_{n+1} = d_n + d_{n-1}$ ($n > 1$). The d_n/d_1 ratios listed in the third column of Table 1.2 are rational numbers whose numerators are given by consecutive *Fibonacci* numbers. These celebrated numbers are defined by the sequence $F_n = \{1, 1, 2, 3, 5, 8, 13, 21, \dots\}$, with $F_0 = F_1 = 1$, where each number in the sequence is just the sum of the preceding two. Quite remarkably, we can appreciate a close relationship between the corresponding d_n/d_1 values and successive powers (rounded to the second decimal place) of the so-called golden mean $\tau = (\sqrt{5} + 1)/2 = 1.6180339887\dots$, as it is

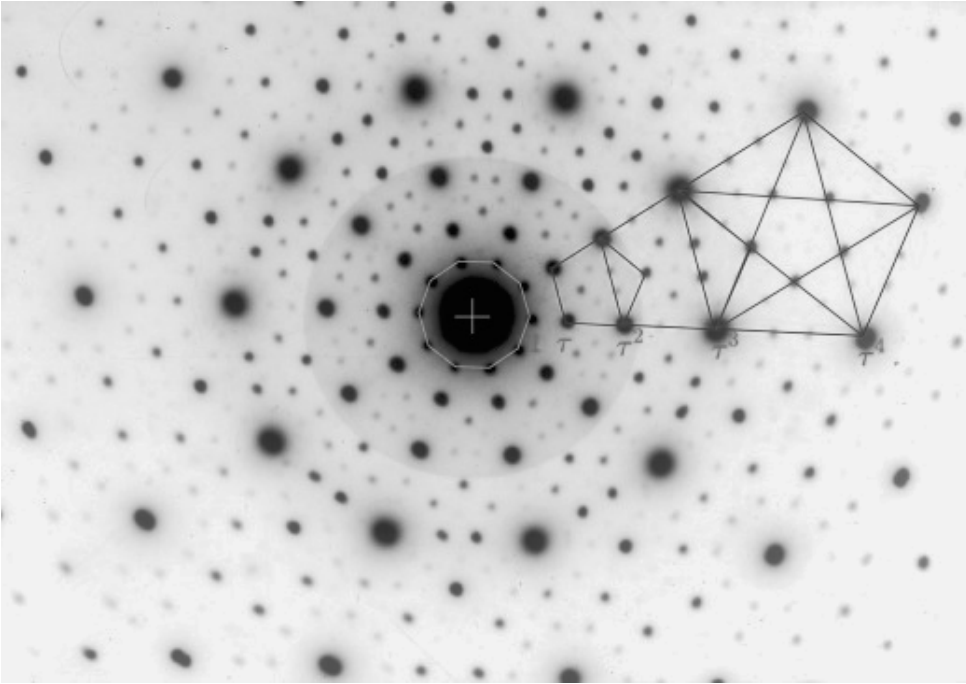


Figure 1.5 Close up view of the electron diffraction pattern shown in Fig. 1.1. In addition to a 10-fold symmetry axis around the origin, a conspicuous series of nested pentagons can be clearly appreciated. Two of them are highlighted at the upper right corner, displaying the so-called Pythagorean pentagram design, obtained by connecting all the opposite vertices by straight lines. The resulting nested pentagons exhibit a size ratio given by τ^{-2} . A sequence of diffraction peaks spaced according to a geometric series based on the golden mean (see Table 1.2) is indicated along the meridian radial path. (Adapted from D. Gratias, M. Quiquandon, *Discovery of quasicrystals: The early days*, C. R. Physique 20, 803-816 (2019). doi: 10.1016/j.crhy.2019.05.009. CC BY-NC-ND license 4.0)[307].

seen by comparing the data listed in the fourth and fifth columns of Table 1.2, respectively. This result indicates that the successive interplanar distances in the considered QC atomic distribution increase by the golden ratio factor.

The golden mean has been largely known from ancient times, since it frequently appears in pentagon and decagon based polygons and polyhedra. This celebrated ratio can be derived in several ways. In general, a segment is said to be divided in the golden mean if the ratio of the whole segment to the larger part is equal to the ratio of the larger to the smaller one. If we take the smaller segment as unit and label the larger part as the unknown x , this geometrical definition can be expressed as $(x + 1)/x = x/1$, which leads to the algebraic equation $x^2 = x + 1$, whose positive solution is the irrational number $x_+ = (\sqrt{5} + 1)/2 = \tau$. Therefore, we get the following expressions relating the golden mean, its square and its reciprocal, namely, $\tau^2 = \tau + 1$, and $\tau^{-1} = \tau - 1$. By successively multiplying the basic

TABLE 1.2 The main Bragg reflections along the meridian radial direction in the diffraction pattern shown in Fig. 1.5 can be labeled according to a power series related to the golden mean.

n	d_n (mm)	d_n/d_1	d_n/d_1	τ^{n-1}	$ \Delta $	(F_n, F_{n-1})
1	7.5	1	1.00	1.00	—	—
2	12.5	5/3	1.67	1.62	0.05	(1, 0)
3	20.0	8/3	2.67	2.62	0.05	(1, 1)
4	32.5	13/3	4.33	4.24	0.09	(2, 1)
5	52.5	21/3	7.00	6.85	0.15	(3, 2)

relation $\tau^2 = \tau + 1$ by τ we get,

$$\begin{aligned}
 \tau^3 &= \tau^2 + \tau = 2\tau + 1, \\
 \tau^4 &= 2\tau^2 + \tau = 3\tau + 2, \\
 \tau^5 &= 3\tau^2 + 2\tau = 5\tau + 3, \\
 &\vdots \\
 \tau^n &= \tau F_{n-1} + F_{n-2}, \quad (n \geq 2).
 \end{aligned} \tag{1.3}$$

Thus, any power of τ can be expressed as a linear combination of two successive Fibonacci numbers in the base $\{\tau, 1\}$. In this way, making use of Eq. (1.3), one can properly label the different diffraction reflections along the radial directions of the diffraction pattern shown in Fig. 1.5 in terms of a pair of Fibonacci numbers, as it is shown in the last column of Table 1.2.

Another useful mathematical relationship between the Fibonacci series and the golden mean is given by the asymptotic limit,⁵

$$\lim_{n \rightarrow \infty} \frac{F_n}{F_{n-1}} = \tau, \tag{1.4}$$

so that the ratio of two successive larger and larger Fibonacci numbers comes closer and closer to the golden mean. If one considers the ratio between next-neighbors in the Fibonacci series one gets,

$$\lim_{n \rightarrow \infty} \frac{F_{n+1}}{F_{n-1}} = \lim_{n \rightarrow \infty} \frac{F_n + F_{n-1}}{F_{n-1}} = 1 + \lim_{n \rightarrow \infty} \frac{F_n}{F_{n-1}} = 1 + \tau = \tau^2. \tag{1.5}$$

By proceeding in a similar way, one can readily see that higher powers of the golden mean are related to asymptotic limits involving ratios of Fibonacci numbers further and further apart along the series. This property is illustrated in the third column of Table 1.2.

At this point, it is important to highlight that the presence of scale invariance symmetry in iQCs can also be observed by studying the diffraction patterns taken along 2- and 3-fold symmetry axes, as it can be readily checked by measuring the distances between successive diffraction peaks along the dashed lines in Fig. 1.4a and Fig. 1.4b, respectively. In doing so, one obtains the same scale factor τ that we obtain when considering the diffraction pattern along the 5-fold axis shown in Fig. 1.4c. This result indicates that the scale invariance symmetry is an isotropic property of iQCs. Therefore, in iQCs the translation symmetry, characteristic of periodic crystals, is *replaced* by an inflation/deflation symmetry

⁵Discovered in 1611 by Johannes Kepler (1571–1630).

in 3D, which is measured in terms of a characteristic scale factor given by the golden mean. The same scale factor is obtained in the case of decagonal QCs (**Exercise 1.2**), whereas for octagonal and dodecagonal QCs one obtains the scale factors given by the irrational numbers $1 + \sqrt{2}/2$ and $2 + \sqrt{3}$, respectively (**Exercise 1.3**). We must note, however, that irrational numbers are beyond the reach of experimental measurement by definition. In fact, the analysis of the electron diffraction patterns performed in the above referred exercises, where all the obtained data can be expressed in terms of rational ratios (see the third column in Tables 1.2, 1.5, 1.6, and 1.7), clearly illustrates this point. Therefore, in order to justify the true irrational nature of the involved scale factors one must rely on some kind of mathematical construction.

The inflation symmetry scale factor can also be derived by constructing the Pythagorean pentagram depicted in Fig. 1.5, taking into account that the ratio between the diagonal and the side of a given pentagon is τ (**Exercise 1.4a**). To this end, one connects all the vertices of a regular pentagon by diagonals. At their intersecting points these diagonals form a smaller pentagon, and the diagonals of this pentagon will form a new pentagram enclosing a yet smaller pentagon. This progression can be continued *ad infinitum*, creating smaller and smaller pentagons and pentagrams in an endless succession exhibiting a self-similar nesting typical of fractal geometry (**Exercise 1.4b**). This self-similar property leads to the appearance of a *hierarchy* of Bragg reflections intensities rendering highly dense diffraction patterns. Indeed, in the ideal case of a perfect QC structure, characterized by the presence of exact irrational scale factors, the distribution of peaks will densely fill the reciprocal space. In actual samples, however, most of the weakest intensity peaks will become extremely dim, making it possible to distinguish individual Bragg spots experimentally. In this way, attending to the relative intensities of the reflections in the diffraction pattern one can readily appreciate the existence of a set of spots which dominate the diffraction pattern, and can be taken to construct a quasi-Brillouin zone. Nevertheless, due to the incommensurate nature of the scale factor we cannot use these peaks to define an average structure compatible with any sort of translation symmetry.⁶

At this point, it is worth mentioning that mineralogists and materials scientists have discovered materials containing icosahedral building blocks, either isolated or linked, well before the discovery of QCs, but these materials exhibited symmetries compatible with periodic order at a large enough scale, as it occurs in fullerite molecular crystal [921], or in organic crystals composed of icosahedral virus particles studied to date (see Sec. 6.1.4.5). In fact, evidence of icosahedral symmetry in condensed matter was early reported in the study of the geometry of virus capsids from X-ray diffraction measurements of several virus crystals. Arguments supporting the possible presence of a high symmetry degree in virus capsids were put forward by Francis H. C. Crick (1916–2004) and James D. Watson, who in 1956, suggested that most of them could be built up from a relatively small number of protein subunits arranged in a symmetrical way, and noticed that of all the possible types of symmetry arrangements only the cubic point groups were likely to lead to an isometric structure [144]. The first experimental evidence for icosahedral symmetry in a virus came from the X-ray diffraction studies of Donald L. D. Caspar on tomato bushy stunt virus [112], soon followed by that of Aaron Klug (1926–2018) and Rosalind E. Franklin (1920–1958) on turnip yellow mosaic virus [428, 429]. These investigations confirmed that the X-ray diffraction patterns from certain virus crystals showed intensity distributions characteristic of a *periodic* arrangement of *icosahedral particles* throughout the space.⁷

⁶At variance with the incommensurately modulated phases case. See, for instance, [381].

⁷For this reason, these virus particles are generally known as icosahedral viruses. Aaron Klug was awarded

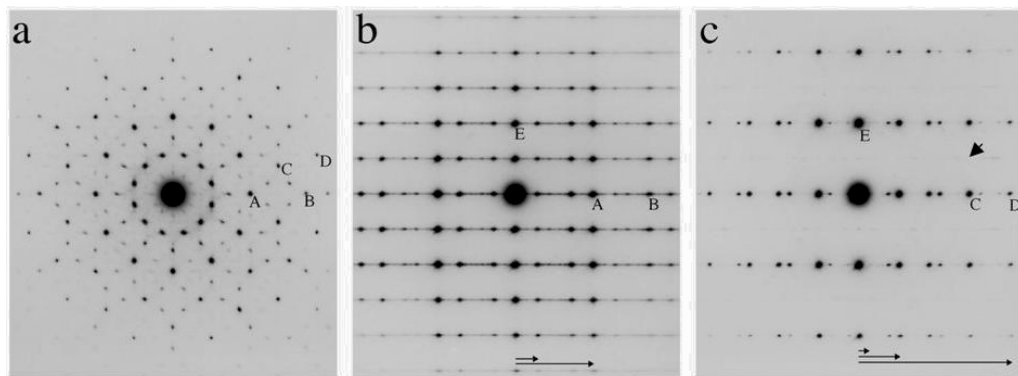


Figure 1.6 Electron diffraction pattern corresponding to a $\text{dd-Mn}_{72.0}\text{Si}_{17.5}\text{Cr}_{5.5}\text{Ni}_{5.0}$ QC taken along the 12-fold axis (a), and along two different kinds of perpendicular 2-fold axes (b) and (c). Arrows at the bottom of (b) and (c) pictures illustrate the characteristic $2+\sqrt{3}$ scaling factor of this axial QC (**Exercise 1.3**). (Taken from Dodecagonal quasicrystal in Mn-based quaternary alloys containing Cr, Ni and Si, Iwami S and Ishimasa T, *Philosophical Magazine Letters*, 85, 229–236, 2005. Reprinted by permission of Taylor & Francis Ltd., <http://www.tandfonline.com>).

Therefore, neither the presence of icosahedral symmetry alone (previously observed in virus assemblies), nor the presence of a scale invariance symmetry (also present in fractal structures) is specific of iQCs. It is the *simultaneous* presence of both types of symmetries in the same piece of matter which makes the difference. Accordingly, in order to properly understand the origin of specific physical properties of QCs, one must properly take into account the characteristic scaling invariance and point group symmetries altogether, introducing a geometrical picture able to blend inflation–deflation relations with rotational and mirror symmetry elements [374].

1.1.3 The sequel: axial quasicrystals

Shortly after the publication of the two seminal papers by D. Shechtman, I. Blech, D. Gratias and J. W. Cahn, and by D. Levine and P. J. Steinhardt, respectively, a number of works reporting the existence of new QCs characterized by the presence of different types of non-crystallographic symmetry axes were published in quick succession. Thus, a new ordered state of matter exhibiting a 12-fold symmetry axis, referred to as dodecagonal QC (ddQC) was found in small particles of a Ni–Cr alloy by T. Ishimasa, H. U. Nissen, and Y. Fukano on 25 July 1985 [367]. An example of the corresponding electron diffraction patterns is shown in Fig. 1.6 for a recently synthesized ddQC in the Mn–Si–Cr–Ni quaternary alloy phase.

Two months later, L. Bendersky reported on another non-crystallographic phase, characterized by the presence of a 10-fold symmetry axis, in rapidly solidified Al–Mn alloys with higher manganese content (18–22 at.%) than those originally studied by Shechtman (10–20 at.%), which was referred to as decagonal QC (dQC) [50, 269]. An example of the corresponding electron diffraction pattern along a 10-fold axis is shown in Fig. 1.7 for a sample

the Nobel Prize in Chemistry, in 1982, for his “development of crystallographic electron microscopy and his structural elucidation of biologically important nucleic acid-protein complexes”.

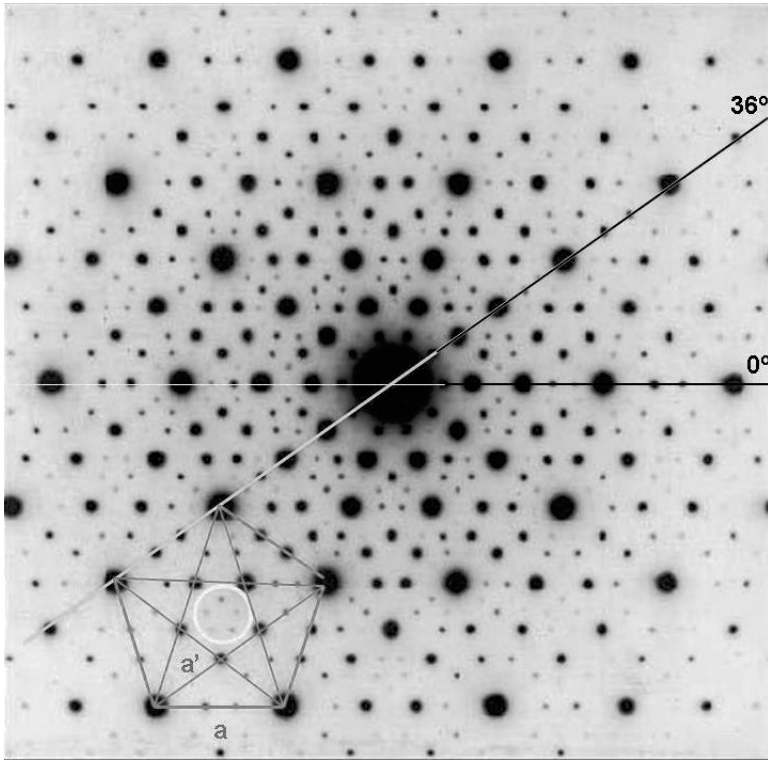


Figure 1.7 Electron diffraction pattern corresponding to a d-Al₇₀Cu₁₅Co₁₅ QC (http://www.tohoku.ac.jp/en/research/research_highlights/images/rh07_01.jpg). A Pythagorean pentagram formed by a large pentagon of side a and a smaller pentagon of side a' is highlighted at the left lower corner. A second generation smaller pentagon is encircled in the pentagram's central region.

belonging to the ternary Al-Cu-Co alloy.⁸ Finally, in 1987, N. Wang, H. Chen, and K. H. Kuo reported the existence of QCs with 8-fold rotational axis in rapidly solidified V-Ni-Si and Cr-Ni-Si alloys, which were referred to as representatives of the octagonal QC class (oQC) [950].

Octagonal, decagonal, and dodecagonal QCs share a common structural design: they consist of transversal planes where the atoms are quasiperiodically arranged displaying 8-, 10-, or 12-fold symmetry axes, respectively, and these planes pile up along the non-crystallographic axis leading to octagonal and dodecagonal platelets or decagonal prisms growth morphologies, as that shown in Fig. 1.8a. Accordingly, the representatives of these three QC classes can be generically referred to as *axial* QCs.

In these materials, two kinds of long-range orders, namely, periodic and QP, coexist in the same sample, at variance with iQCs which display quasiperiodic order (QPO) all

⁸It is interesting to note that the observation of a diffraction pattern with a pseudo-pentagonal symmetry had earlier been reported in an investigation by transmission electron microscopy (the same technique used by Shechtman) of rapidly solidified AlPd alloys in 1978. Unfortunately, not enough attention was paid to this system, which was later recognized as belonging to the decagonal phase [210].

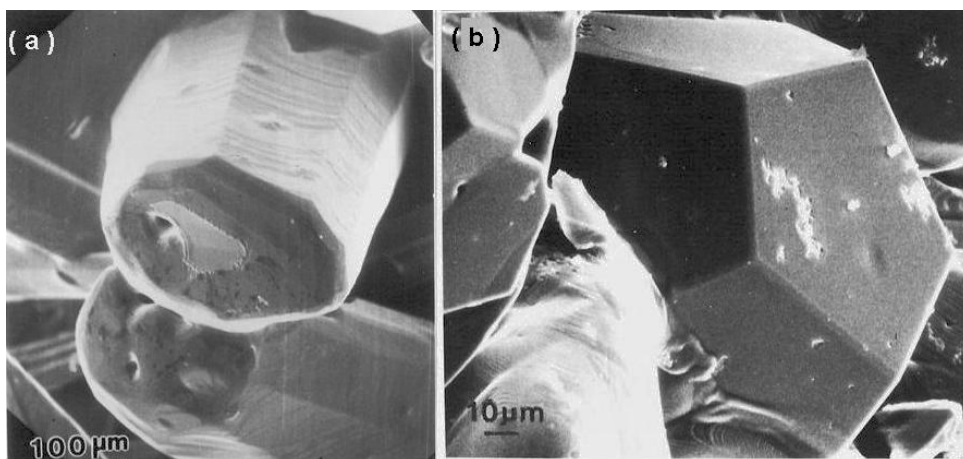


Figure 1.8 Scanning electron images showing the growth morphology of two representative thermodynamically stable QCs. (a) A decagonal prism habit corresponding to the d-AlNiCo QCs. (b) A regular pentagonal dodecahedral habit corresponding to the i-AlCuFe QCs. (Courtesy of An Pang Tsai).

over the space, exhibiting dodecahedral (Fig. 1.8b) or triacontahedral (Fig. 1.9) growth morphologies. This interesting feature suggested that other kinds of *mixed order* structures may also exist. In fact, a one-dimensional QC derived from decagonal phase representatives was found in rapidly solidified AlNiSi, AlCuMn, and AlCuCo alloys. In this class of QCs, in addition to the periodic translation along the 10-fold axis present in axial QCs, the translation along one of the 2-fold axes normal to the 10-fold one becomes periodic as well [329].

In Fig. 1.9, we present a classification of QCs attending to the number of spatial dimensions in which the QPO occurs. For any given solid its 3D spatial dimension is expressed as the sum of its periodic dimension (labelled by the corresponding row number) plus its QP dimension (labelled by the corresponding column number). In this way, the coordinates of a given solid in the chart indicate the relative importance of QP versus periodic order in its structure. For example, the iQC shown at the left upper corner exhibits QPO along the three dimensions of space, so that it can be regarded as an example of isotropic QC. On the contrary, dQCs located at the middle of the chart exhibit QPO in the planes perpendicular to the decagonal axis, but are periodically arranged along this axis, so that they must be regarded as a representative example of anisotropic QCs. At the right lower corner we have another interesting instance of a hybrid-order system: a Fibonacci heterostructure. In this system a series of periodic crystal layers are stacked following a QP 1D array. As we will see in Sec. 4.2.1, this kind of structures have been artificially grown and they properly illustrate the technological potential related to the very notion of hybrid-order QP designs in materials science.

1.1.4 Thermodynamically stable quasicrystals grow large

“At the end of the 18th century, it was realized that the shape of natural crystals and the angles between exposed facets reflected their internal symmetry.

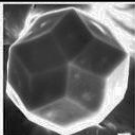
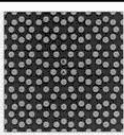
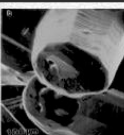
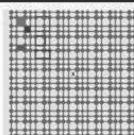
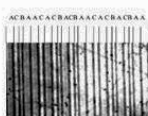
QUASICRYSTALS CLASSIFICATION CHART				
		QUASIPERIODIC ORDER DIMENSIONALITY		
PERIODIC ORDER DIMENSIONALITY		3D	2D	1D
	0D			
	1D			
	2D			

Figure 1.9 Classification of different types of QCs attending to their QP dimensionality. From left to right and from top to bottom we see a triacontahedral iQC (<https://blog.stephenwolfram.com/data/uploads/2018/12/quasicrystals-rhombic-triacontahedra-2.png>), a photonic oQC, a dQC, a Fibonacci code bar[246], a Fibonacci-periodic hybrid square, and a dielectric Fibonacci multilayer (Adapted from Maciá E, The role of aperiodic order in science and technology, Rep. Prog. Phys. 69, 397–441, 2006; doi:10.1088/0034-4885/69/2/R03 © IOP Publishing. Reproduced with permission. All rights reserved).

Similarly, faceted samples have been observed in quasicrystalline materials grown in the laboratory”. (Julian Ledieu and Vincent Fournée, 2014) [483].

First generation (1984–1986) QCs were obtained by means of far from equilibrium techniques and exhibited small sizes, typically a few micrometers, which is too small to precisely investigate their intrinsic physical properties. In fact, they were not even thermodynamically stable and transformed into periodic crystals after heating at moderate temperatures (Fig. 1.10). In addition, their X-ray diffraction lines were rather broad, resembling those observed in highly faulted periodic crystals. Thus, during the early years of the field, it was naturally assumed that QCs were located at a somewhat intermediate position between periodic crystals and amorphous materials, so that they should be inherently unstable. This assumption proved, however, to be wrong.

In 1955, H. K. Hardy and J. M. Silcock classified the intermetallic compounds in equilibrium with the aluminum solid solution in the Al-Li-Cu system by using a Debye-Sherrer power diffraction method. At 500°C, they found several compounds corresponding to hexagonal and cubic structures, in addition to a phase they called T2, with composition close to $\text{Al}_6\text{Li}_3\text{Cu}$, which exhibited a X-ray diffraction spectrum escaping any crystalline indexation

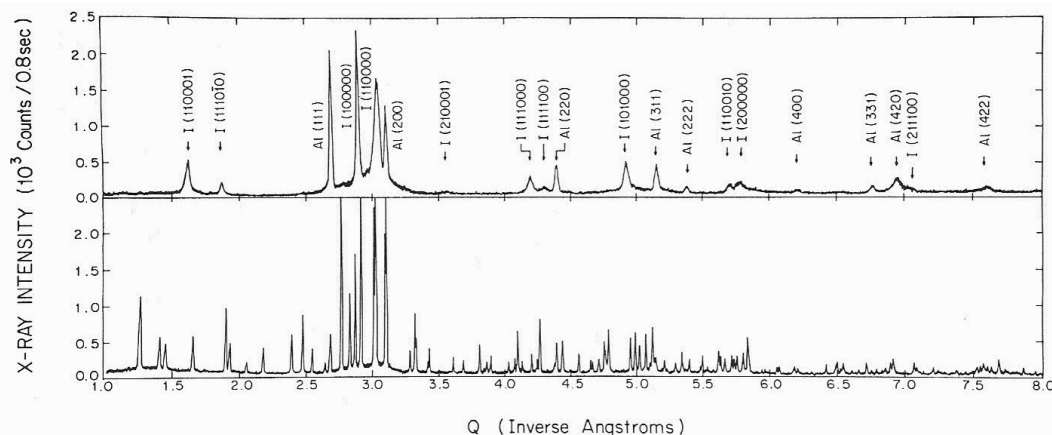


Figure 1.10 (Top panel) High-resolution X-ray diffraction pattern of quenched $i\text{-Al}_{86}\text{Mn}_{14}$ quasicrystalline powder. Some contaminating fcc Al phase is present. Icosahedral phase peaks are labeled using generalized Miller indices in 6D. (Bottom panel) Diffraction pattern of annealed $i\text{-Al}_{86}\text{Mn}_{14}$ powder by heating to 410°C for 75 minutes in an argon atmosphere. All peaks can be indexed as belonging to the Al_6Mn orthorhombic phase. (Reprinted figure with permission from Bancel P A, Heiney P A, Stephens P W, Goldman A I, and Horn P M, *Physical Review Letters* 54, 2422, 1985. Copyright (1985) by the American Physical Society).

[316, 320]. This oddity remained dormant for more than three decades, until B. Dubost, J. M. Lang, M. Tanaka, P. Sainfort, and M. Audier proved that this phase actually belongs to the iQC class. In a paper entitled “*Large AlCuLi single quasicrystals with triacontahedral solidification morphology*”, published in *Nature* on 6 November 1986, they announced the discovery of the first thermodynamically stable iQC, thus being able to grow large grains up to the millimeter size by conventional solidification techniques in close to equilibrium conditions [213]. In this way, large quasicrystalline grains, exhibiting the growth morphology of a multi-faceted rhombic triacontahedron (Fig. 1.9 left top panel), were demonstrated in $i\text{-Al}_{56}\text{Li}_{33}\text{Cu}_{11}$ QCs by several groups. Such a triacontahedral morphology was a novelty in crystallography and mineralogy.

Shortly after, two new stable iQC representatives, now exhibiting pentagonal dodecahedral solidification morphologies, were reported by A. P. Tsai, A. Inoue, and T. Masumoto in the $i\text{-Al}_{65}\text{Cu}_{20}\text{Fe}_{15}$ QC (September 1987) [904], and by W. Ohashi and F. Spaepen in $i\text{-Zn}_{43}\text{Mg}_{37}\text{Ga}_{20}$ QC (December 1987) [660], respectively.⁹

The structural quality of a sample can be estimated from the X-ray diffraction spectra in terms of the so-called correlation length, given by the relation $2\pi/\text{FWHM}$, where FWHM is the diffraction peak full width at half maximum. According to this criterion, first generation QCs did not possess a very good structural quality, showing correlation lengths in the approximate range 50–100 nm. During the period 1987–2000, a remarkable number of thermodynamically stable iQCs of high structural quality were discovered by A. P. Tsai and co-workers in the $\text{AlCu}(\text{Fe}, \text{Ru}, \text{Os})$, $\text{AlPd}(\text{Mn}, \text{Re})$, $\text{Zn-Mg}(\text{Y}, \text{Gd}, \text{Tb}, \text{Dy}, \text{Ho}, \text{Er})$,

⁹For the sake of historical curiosity both manuscripts were received on 14 July 1987 in their respective journals.

TABLE 1.3 Main representatives of thermodynamically stable iQCs [840, 502, 68, 145, 297, 462, 874]. RE stands for rare-earth atoms. The star marked sample corresponds to the mineral QC icosahedrite (see Sec. 1.2).

Al-based	Zn-based	Ti-based	Cd-based	Ag/Au-based
Al _{70.5} Pd ₂₁ Mn _{8.5}	Zn ₈₈ Sc ₁₂	Ti ₄₅ Zr ₃₈ Ni ₁₇	Cd _{88.3} RE _{11.7}	Ag _{62.7} Al ₂₇ Ca _{14.3}
Al ₇₀ Pd ₂₀ Re ₁₀	Zn ₈₄ Ti ₈ Mg ₈	Ti ₄₀ Hf ₄₀ Ni ₂₀	Cd ₈₅ Yb ₁₅	Au _{60.0} Sn _{26.7} Yb _{13.3}
Al ₆₅ Cu ₂₅ Fe ₁₅	Zn ₈₀ Sc ₁₅ Mg ₅		Cd ₈₅ Ca ₁₅	Au ₄₆ Al ₃₈ Tm ₁₇
Al ₆₅ Cu ₂₅ Ru ₁₅	Zn ₇₇ Sc ₁₅ Mg ₅		Cd _{88.3} RE _{11.7}	Ag _{44.2} In _{41.7} Ca _{14.1}
Al ₆₅ Cu ₂₅ Os ₁₅	Zn ₇₅ Sc ₁₆ Fe ₇		Cd _{66.8} Mg _{21.4} RE _{11.8}	Ag ₄₂ In ₄₂ Yb ₁₆
*Al ₆₃ Cu ₂₄ Fe ₁₃	Zn _{71.5} Sc _{16.2} Cu _{12.3}		Cd ₆₅ Mg ₂₀ Ca ₁₅	Au ₄₂ In ₄₂ Ca ₁₆
Al ₅₆ Li ₃₃ Cu ₁₁	Zn ₆₀ Mg ₃₀ RE ₁₀		Cd _{54.6} Mg _{33.0} RE _{12.4}	
Al ₅₄ Pd ₃₀ Sc ₁₆	Zn ₄₃ Mg ₃₇ Ga ₂₀			

TABLE 1.4 Main representatives of thermodynamically stable dQCs attending to the number of layer periods along the 10-fold axis [490, 502, 661, 840]. RE stands for rare-earth atoms. The star marked sample corresponds to the mineral QC decagonite (see Sec.1.2).

2	4	6	8
*Al ₇₁ Ni ₂₄ Fe ₅	Al ₇₀ Ni ₁₅ Fe ₁₅	Al _{70.5} Mn _{16.5} Pd ₁₃	Al ₇₅ Pd ₁₅ Os ₁₀
Al ₇₀ Ni ₁₅ Fe ₁₅	Al ₇₀ Ni ₂₀ Rh ₁₀	Al ₆₅ Rh ₂₀ Cu ₁₅	Al ₇₃ Ir _{14.5} Os _{12.5}
Al ₇₀ Ni ₂₀ Rh ₁₀	Al ₆₅ Cu ₂₀ Co ₁₅	Co ₄₇ Ga ₄₃ Cu ₁₀	Al ₇₀ Ni ₂₀ Ru ₁₀
Al ₆₅ Rh ₂₀ Cu ₁₅	Al ₆₅ Cu ₂₀ Ir ₁₅		
Mn _{47.4} In _{37.8} Ni _{14.8}	Al ₆₅ Rh ₂₀ Cu ₁₅		
In _{44.6} Ni _{27.2} Mn _{23.2}	Zn ₆₈ Mg ₄₀ RE ₂		

and Cd(Yb,Ca) alloy systems [904, 906, 908]. A number of thermodynamically stable dQCs was also discovered by this team in the AlCo(Cu,Ni) alloy system [905]. In 2005, An Pang Tsai (1958–2019) received the Jean Marie Dubois Award “*in recognition of fundamentally important discoveries of new quasicrystalline phases*”. On the other hand, new thermodynamically stable iQCs were reported by K. F. Kelton, Y. J. Kim, and R. M. Stroud in the Ti-Zr-Ni alloy system in 1997 [409], and by John D. Corbett (1926–2013) and Q. Lin in the Zn-Sc-Cu and (Au/Ag)-In-(Ca/Yb) systems in 2003 and 2007, respectively. The first stable dodecagonal phase, dd-Ta₆₂Te₃₈, was discovered in 1998 by M. Conrad, F. Krumeich, and B. Harbrecht. Currently, about two hundred QC representatives belonging to a broad variety of alloy systems have been reported to be thermodynamically stable up to their respective melting points and to exhibit diffraction patterns of extraordinary quality, some of them comparable to those observed in the best monocrystalline samples ever grown. For the sake of information some representative compounds belonging to the iQC and dQC classes are listed in Tables 1.3 and 1.4, respectively. With a few exceptions most stable dQCs are Al-based and they are classified attending to the number of atomic layers (i.e., 2, 4, 6, 8) in the period along the 10-fold axis. The growth of large size, high structural quality stable QC samples allowed for the study of the physical properties of these compounds, which we will discuss in detail in Chapter 3.

1.1.5 Revamping the very crystal notion

The discovery of QCs was a terrible shock for the crystallographical, solid state, and condensed matter communities. Indeed, quasicrystalline alloys were unexpectedly found, once the theoretical framework aimed to understand the nature of crystal kingdom from a

microscopic viewpoint was regarded to be essentially completed on rigorous mathematical grounds, within a conceptual scheme which had proved very successful in accounting for the structure of every form of matter studied during seven decades. And such a rigorous and successful theory explicitly forbade the growth morphologies and atomic arrangements exhibited by icosahedral, decagonal, octagonal, and dodecagonal QCs!

This apparent paradox naturally spurred a hectic period of intellectual storm, and the solution fortunately arrived relatively soon. It turned out to be quite simple in retrospective: our previous theoretical understanding of ordered condensed matter was entirely based on the very notion of periodic arrangements of atoms in space. But what about aperiodic ones? We had simply missed them in constructing classical crystallography.

“Because no exceptions were found in almost 200 years, it became accepted that all crystals were regular arrays on a lattice, and became a law or paradigm that arose from experience, rather than from fundamental principles”. (John Werner Cahn, 2014) [104]

Consequently, all the conceptual troubles stemming from the discovery of this new form of matter were rooted on an epistemological shortcoming rather than pointing to any sort of pathological state of matter. The message from nature was that matter can display well-ordered arrangements well beyond the rules imposed by strict periodicity, thereby lifting the restrictions regarding the rotational symmetries that can leave the atomic order invariant. In this way, the puzzle originally risen by the crystallographical restriction theorem was eventually clarified, allowing for new progresses in our understanding of the atomic arrangements in solids by the light of the notion of *aperiodic crystal*, as it was heralded by E. Schrödinger (1887–1961):

“In physics we have dealt hitherto only with periodic crystals. To a humble physicist’s mind, these are very interesting and complicated objects; they constitute one of the most fascinating and complex material structures by which inanimate nature puzzle his wits. Yet, compared with the aperiodic crystal, they are rather plain and dull”. (Erwin Schrödinger, 1944) [789]

Accordingly, the International Crystallographic Union approved in April 1991, the establishment of a *Commission on Aperiodic Crystals* with the membership of J. M. Pérez-Mato (Chairman), G. Chapuis, M. Farkas-Jahnke, M. L. Senechal, and W. Steurer. According to the terms of reference introduced by this Commission:

In the following 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 [357].

Thus, QCs belong to the novel aperiodic crystals category,¹⁰ whereas usual periodic crystals are now known as *classical* crystals. The revamped crystal definition reflects our current understanding that periodicity at the atomic scale is a sufficient but not necessary condition for crystallinity. Instead, the presence of a long-range atomic order *able to diffract* must be regarded as the essential attribute of crystalline matter rather than mere periodicity. Consequently, within the crystalline family we can now distinguish between periodic crystals, which display periodic arrangements of atoms, and aperiodic crystals, lacking such

¹⁰Along with incommensurate modulated crystals and incommensurate composites [381].

a periodicity, which is replaced by other kinds of symmetries, such as scale invariance (inflation symmetry) or long-range repetitiveness. Thus, the existence of a mathematically well defined long-range atomic order should be properly regarded as the *generic* attribute of ordered solid state matter.

In this context, it is convenient to highlight that, although both random structures and aperiodically ordered lattices lack strict translational symmetry, the *absence of periodicity* characteristic of aperiodic systems (e.g., QCs, incommensurate phases, or fractal structures) are not the same in nature as the *complete lack of long-range order* characteristic of amorphous matter. In fact, as we mentioned above, in aperiodic systems translational symmetry is replaced by other symmetry properties which amorphous matter does not possess.

The importance of the scale invariance property in defining the very QC notion is illustrated by the fact that one can consider the existence of QCs exhibiting rotational symmetries which are *allowed* by the classical restriction theorem (namely, 2-, 3-, 4-, and 6-fold). In fact, albeit the most striking symmetry feature of QCs at their discovery was the presence of 5-, 8-, 10-, or 12-fold rotation axes in their diffraction patterns, it is natural to question as to whether this is an essential condition for a solid to be properly considered as a QC. It is currently agreed that the presence of non-crystallographic axes is not a necessary condition for a solid to be regarded as a QC, and that the key feature to this end is just to exhibit scale invariance symmetry [381]. Indeed, several examples of QCs exhibiting 2-, 3-, and 4-fold symmetry axes *along with* scale invariance symmetry characterized by *irrational* scale factors have been reported, being referred to as “cubic QCs” [229]. These findings support the view that QC definition *should not include* the requirement that they must display a classically *forbidden* axis of symmetry, as it is stated in the *Online Dictionary of Crystallography* of the International Union of Crystallography, where one reads:

*Often, quasicrystals have crystallographically ‘forbidden’ symmetries (...). However, the presence of such a forbidden symmetry is not required for a quasicrystal.*¹¹

An illustrative example of 2D QC without forbidden symmetries is the so-called square Fibonacci tiling, with tetragonal point group symmetry (4 mm) [494]. This square can be generated by superposition of two Fibonacci line grids, which are orthogonal to each other. In this way we obtain an infinite set of lines whose inter-line spacing follows the Fibonacci sequence of short (unity) and long (τ) distances. The resulting two-dimensional QP structure is shown in Fig. 1.11a, and it consists of three different tiles: (a) a large square of dimensions $\tau \times \tau$, (b) a small square of dimensions 1×1 , and (c) a rectangle of dimensions $1 \times \tau$ (see **Exercise 2.7**). A similar construct can be extended to obtain a cubic Fibonacci tiling in 3D (see Fig. 4.9) [579]. An experimental realization of the Fibonacci square grid structure was obtained by selectively depositing C₆₀ fullerene molecules atop Mn atoms on the 2-fold surface of an i-AlPdMn QC, as it is shown in Figs. 1.11b–c [136].

1.1.6 The reciprocal space take-over

“It was a misfortune of intellectual history that the space-group classification scheme of crystallography was developed in real space to categorize periodic structures, rather than in Fourier space to categorize diffraction patterns consisting of sharp Bragg peaks”. (N. David Mermin, 1992) [600]

¹¹<https://reference.iucr.org/dictionary/Quasicrystal>. Last accessed on 10 January 2020. Notwithstanding this, it is fair to say that some authors consider that the term QC should denote crystals with diffraction patterns showing non-crystallographic symmetry axes in order to distinguish them from other representatives belonging to the aperiodic crystals class [840].

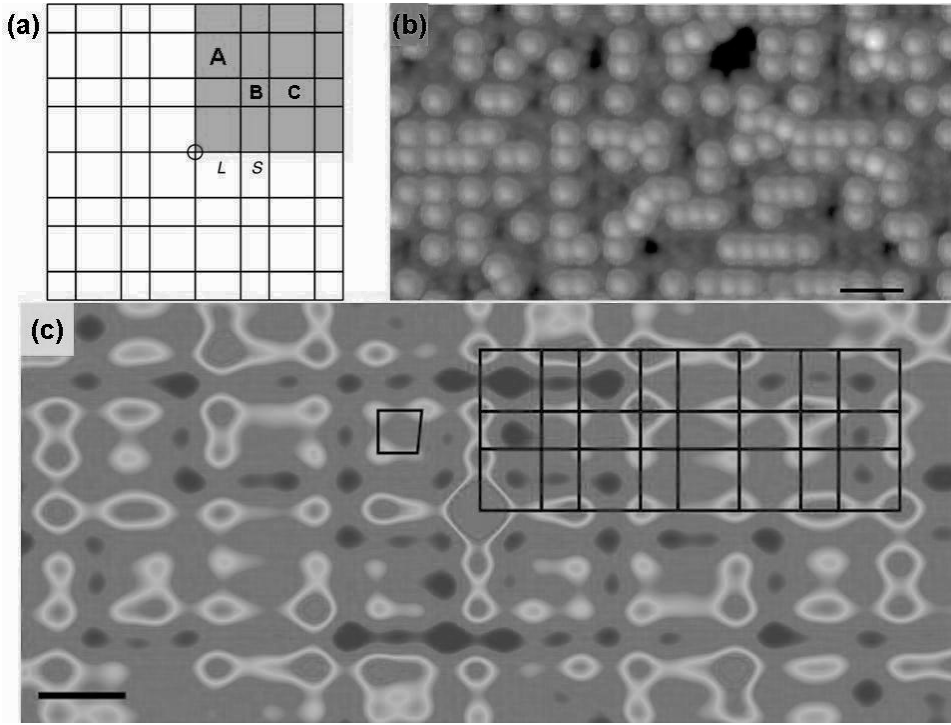


Figure 1.11 (a) The Fibonacci square grid with the constituent A, B, and C tiles. The center of rotation of 4-fold symmetry is marked by a circle. (b) Fullerene molecules deposited atop Mn atoms on the 2-fold surface of an i-AlPdMn single grain QC imaged by scanning tunneling microscopy. (c) Autocorrelation function of the C_{60} overlayer shown in (b). A distorted square is marked along with the Fibonacci square grid with $L = 2.04$ nm and $S = 1.26$ nm ($L/S = 34/21 \simeq 1.619$). (Adapted from Coates S, Smerdon J A, McGrath R, Sharma H R, A molecular overlayer with the Fibonacci square grid structure, *Nature Commun.* 9 3435 (2018), doi:10.1038/s41467-018-05950-7. CC NY-NC-ND license 4.0).

According to its very definition the diffraction pattern of an aperiodic crystal must mainly consist of sharp Bragg reflections, like those of periodic classical crystals, albeit lacking 3D periodicity. Thus, a fundamental aspect of the revamped definition of crystal is that the conceptual focus is shifted from direct space, where the atoms are physically located, to reciprocal space, where their underlying order is unveiled by means of diffraction phenomena.¹² In fact, diffraction patterns are closely related to the Fourier transform of the lattice atomic distribution $\rho(\mathbf{r})$, given by the expression

$$F(\mathbf{k}) = \int_V \rho(\mathbf{r}) e^{i\mathbf{k} \cdot \mathbf{r}} d\mathbf{r}, \quad (1.6)$$

¹²It is likely that the transition from X-ray crystal structure analysis to high resolution electron microscopy as a paradigmatic method for the investigation of crystal structures, had played a significant role in promoting this conceptual shift [568].

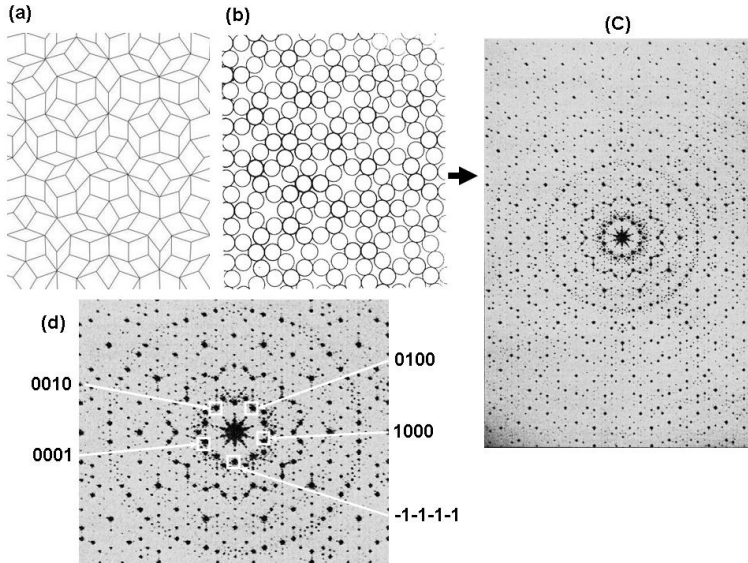


Figure 1.12 (a) A portion of the 2D Penrose tiling made of acute and obtuse rhombi. (b) Arrangement of circles centered at each vertex of a Penrose tiling (c) Optical diffraction pattern of this arrangement made by G. Harburn. The annular objects show in the circular strong and weak modulations of the pattern, which itself exhibits local 10-fold symmetry. (d) Magnification of the central region of the pattern shown in (c) with the indexing of the main spots in terms of four integers. (Reprinted from *Physica A: Statistical Mechanics and its Applications*, 114, Mackay A L, *Crystallography and the Penrose pattern*, 609–613, Copyright (1982), with permission from Elsevier).

where $\mathbf{k}$ denotes reciprocal space vectors and the volume integral extends over all the space. Thus, Fourier transform can be envisioned as a linear transformation relating two different mathematical domains: that corresponding to the space vector $\mathbf{r}$ where the atoms reside, and that corresponding to the $\mathbf{k}$ vectors related reciprocal space. The key question regarding the very nature of possible orderings of matter (either periodic or aperiodic ones) can then be formulated as follows: given a certain arrangement of atoms in terms of a distribution function $\rho(\mathbf{r})$, which are the necessary and sufficient conditions in order for its Fourier transform to exhibit an essentially discrete spectrum?

A significant step forward in order to solve this fundamental issue was given by Alan L. Mackay, who in 1982 reported that a distribution of circles arranged according to the 2D QP Penrose pattern was able to diffract (Fig. 1.12).

“(...) Alan (Mackay) presented his optical diffraction pattern of Penrose tiling vertices, heralding the once-unthinkable, now ever-expanding research field of aperiodic order. A few months later, Dan Shechtman discovered AlMn crystals that had a strikingly Penrose-like diffraction pattern, and crystallography changed for ever”. (Marjorie Senechal, 2017)[796]

The diffraction pattern shown in Fig. 1.12 displays local 10-fold axis, and thus represents a structure outside the formalism of classical crystallography, which was designated as

a *quasi-lattice* by Mackay [566]. The quasilattice notion was introduced as an array of points in *reciprocal space*, where each point has integer indices with respect to a base of vectors and where the base dimension is larger than the dimensionality of the physical space [565]. For instance, in order to index the diffraction pattern obtained by Mackay one needs four integer indices. The four basis vectors are mapped on positions with integer indices under a $2\pi/5$ rotation (note the pentagonal arrangement of the peaks highlighted in Fig. 1.12d), as $\{\cos(2\pi n/5), \sin(2\pi n/5), \cos(4\pi n/5), \sin(4\pi n/5)\}$, $n = 1, \dots, 4$. This gives a four-dimensional matrix, which on an orthogonal basis combines a two-dimensional rotation over $2\pi/5$ with another two-dimensional rotation over $4\pi/5$ [382]. Note that in the term quasilattice the prefix “quasi” is used to design a perfectly ordered structure in higher dimensions, not a defective or flawed one. Thus, although Mackay was completely aware of the perfectly ordered nature of the structure described in terms of the quasilattice notion, he did not use other possible terms, such as “super-lattice” or “hyper-lattice” to this end.¹³ In 2010, Alan L. Mackay was awarded the Oliver E. Buckley Condensed Matter Prize from the American Physical Society for his “*pioneering contributions to the theory of quasicrystals, including the prediction of their diffraction pattern*”.

In 1987, it was noted by Rokhsar, Mermin, and Wright that if a QC diffraction pattern contains reflections corresponding to two wave vectors, say $\mathbf{k}_1$ and $\mathbf{k}_2$, one in general will find reflections corresponding to the sum, $\mathbf{k}_1 + \mathbf{k}_2$, and difference $\mathbf{k}_1 - \mathbf{k}_2$ as well, unless such reflections are forbidden by symmetry constraints. QCs, like periodic crystals, can therefore be indexed by a set of wave vectors that is closed under addition (sums and differences). For a periodic crystal this vector set reduces to the so-called reciprocal lattice, the set of wave vectors of plane waves with the periodicity of the direct lattice in physical space. For a QC this set still forms a lattice (in the sense that it is closed under addition), but unlike a periodic reciprocal lattice a quasicrystalline reciprocal lattice has no minimum distance between points, and the corresponding set of plane waves has no common real-space period. Although quasicrystalline wave vectors form a dense set in $\mathbf{k}$ space, a quasicrystalline diffraction pattern admits a countable indexing, unlike the diffraction pattern of an amorphous material [751].

It is thus reasonable to address the crystallographic classification of QCs from the reciprocal space viewpoint by cataloging the allowable lattices attending to the point group symmetry of the considered QC. These lattices are generated by *six* or *five* vectors for icosahedral and decagonal lattices, respectively, which are derived from the corresponding diffraction patterns. The icosahedral lattice can be defined in terms of a base of dimension 3 and rank 6 given by six vectors in the reciprocal space $\mathbf{a}_i^*$, $i = 1, \dots, 6$, pointing from the center to the non-aligned vertices of an icosahedron (Fig. 1.13). The integral linear combinations of the $\mathbf{a}_i^*$ vectors define the points of the icosahedral lattice

$$\Lambda^* = \left\{ \sum_{i=1}^6 n_i \mathbf{a}_i^* \mid n_i \in \mathbb{Z} \right\}, \quad (1.7)$$

¹³As a matter of fact, the term “super-lattice” had been previously introduced by Leo Esaki and R. Tsu in 1970, in order to describe an artificial periodic structure consisting of alternate layers of two different semiconducting materials, with layer thickness of the order of nanometers [235]. They called this synthetic structure a *superlattice* because periodic order appears at two different scale lengths: at the atomic level, we have the usual crystalline order determined by the periodic arrangement of atoms in each layer (lattice structure), whereas at longer scales, we have the periodic order determined by the sequential deposition of the different layers (superlattice structure).

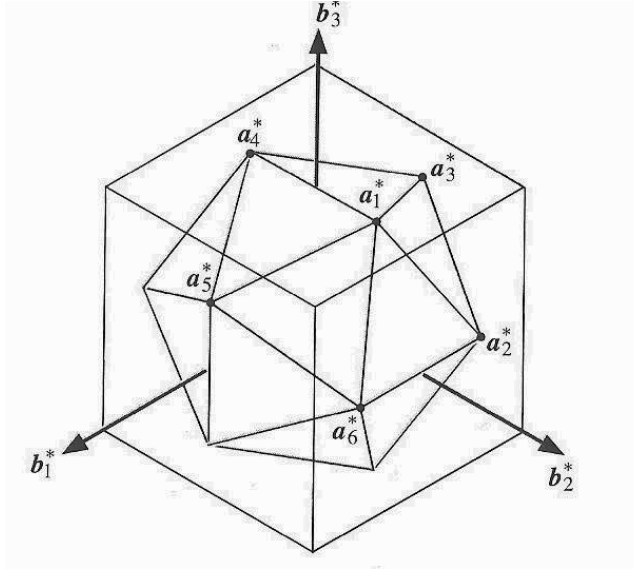


Figure 1.13 Perspective view of the two alternative reciprocal bases generally used to study icosahedral lattices in reciprocal space: the cubic and the icosahedral settings, represented by the bases $\mathbf{b}_i^*$ and $\mathbf{a}_i^*$, respectively. The Cartesian coordinate system is centered at the icosahedron center and oriented along three mutually orthogonal 2-fold axes of the icosahedron.

where the basis vectors $\mathbf{a}_i^*$ are given by

$$\begin{pmatrix} \mathbf{a}_1^* \\ \mathbf{a}_2^* \\ \mathbf{a}_3^* \\ \mathbf{a}_4^* \\ \mathbf{a}_5^* \\ \mathbf{a}_6^* \end{pmatrix} = \begin{pmatrix} 0 & 1 & \tau \\ -1 & \tau & 0 \\ -\tau & 0 & 1 \\ 0 & -1 & \tau \\ \tau & 0 & 1 \\ 1 & \tau & 0 \end{pmatrix} \begin{pmatrix} \mathbf{b}_1^* \\ \mathbf{b}_2^* \\ \mathbf{b}_3^* \end{pmatrix}, \quad (1.8)$$

in the $\mathbb{R}^3$ orthonormal basis $\mathbf{b}_i^*$, $i = 1, \dots, 3$ [840]. Accordingly, the diffraction reflections can be labeled by six generalized Miller indices and the diffraction pattern is spanned by six linearly independent reciprocal lattice vectors. The space group classification of QCs belonging to the icosahedral point groups Y and Y_h within the reciprocal space framework was addressed by David Mermin and co-workers in 1988, who showed that taking into account the scaling properties of QCs is crucial for a correct enumeration of their related space groups [751].

1.2 MINERAL QUASICRYSTALS

“Had we found a crystal? Many definitions of crystals are in use, some have changed over the centuries. Our solid (...) can be grown to form ‘a convex solid enclosed by symmetrically arranged plane surfaces, intersecting at definite and characteristic angles’. According to the latter of these older definitions, quasicrystals are crystals”. (John W. Cahn, 2001) [103]

Carlo or molecular dynamics simulations employing specific pair potentials, though whether periodic or QPO results in a low enough free energy for a given mass density and chemical composition of the melt largely depends on the cluster packing optimization rather than on pair potentials directly [841]. Due to multibody interactions and the existence of itinerant electrons, the pair potentials of atoms in intermetallic phases strongly depend on the kind and arrangement of neighboring atoms. Thus, Al-TM based QCs, where d-orbitals play a crucial role can be described in terms of Friedel oscillations, whereas simpler Lennard-Jones potentials of the form

$$U(r) = U_0 \left[\left(\frac{r_0}{r} \right)^{12} - \left(\frac{r_0}{r} \right)^6 \right], \quad (1.33)$$

where r_0 measures the location of the potential minimum, are appropriate in order to model Mg-Zn based QCs with s- and p-electrons dominating metallic bonding [758]. Local disorder sources, such as phason flips, can be studied quite well in terms of these classical simulation methods, which can deal with system sizes up to millions of atoms. In turn, quantum mechanical first principles calculations can shed light onto the nature of local chemical bonding of energetically favorable, optimal cluster building blocks. Nevertheless, these calculations can only consider model systems with a size below a thousand atoms, which corresponds to just a few clusters per unit cell. Consequently, the role of long-range QPO can not be properly modeled and the information obtained from these calculations refers to low-order approximant model structures rather than to QP ones²² [841].

1.6 EXERCISES WITH SOLUTIONS

1.1 Determine the allowed rotation axes in two-dimensional periodic lattices.

In two dimensions the orthogonal matrix describing rotations by an angle φ reads

$$\mathbf{M} = \begin{pmatrix} \cos \varphi & -\sin \varphi \\ \sin \varphi & \cos \varphi \end{pmatrix}, \quad (1.34)$$

so that the crystallographical restriction in 2D can be analogously written as

$$\text{tr} \mathbf{M} = 2 \cos \varphi \equiv n \in \mathbb{Z}, \quad (1.35)$$

which can be rewritten in the simple form $\cos \varphi = n/2$. The condition $|\cos \varphi| \leq 1$ implies $n = \{-2, -1, 0, 1, 2\}$ in this case. Quite remarkably, however, by plugging these integer numbers in the expression $\varphi = \cos^{-1}(n/2)$, we obtain exactly the same solutions listed in Table 1.1. Therefore, the crystallographical restriction leads to the *same* set of allowed rotation axes for 2D and 3D periodic lattices [32]. This is a general feature of successive $2nD$ and $(2n+1)D$ couples of Euclidean hyperspaces (see [840] for more details).

1.2 Following the procedure described in Section 1.1.2 study the relative distances to the center of the main diffraction peaks along a radial direction for the (a) decagonal, and (b) icosahedral QCs shown in Figs. 1.7 and 1.27, respectively. Compare the obtained results with those reported in Table 1.2.

(a) Firstly, we measure the distances of successive main diffraction spots along the radial direction outwards from the center, d_n , and calculate the ratios d_n/d_1 . The obtained values

²²This shortcoming may be overcome by implementing a suitable renormalization scheme able to exploit the underlying self-similar structure of full-fledged QCs.

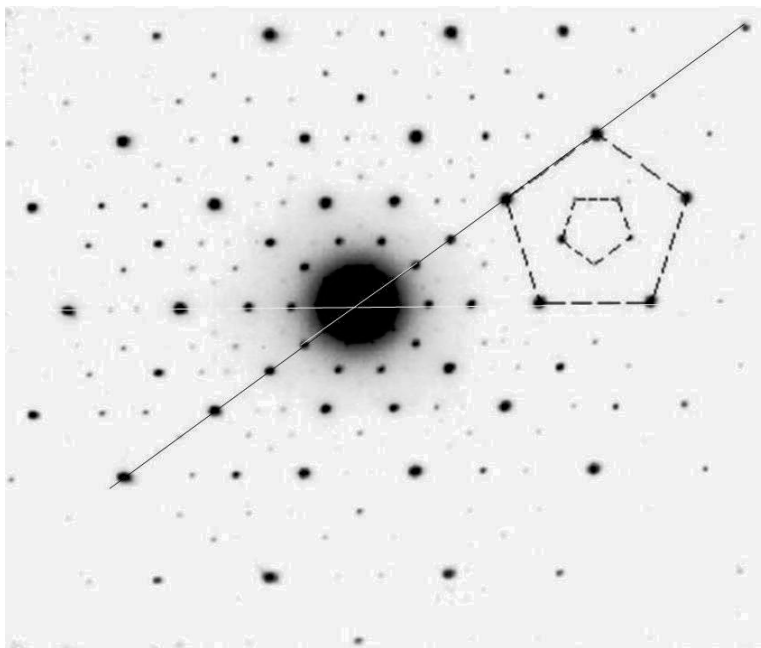


Figure 1.27 Electron diffraction pattern corresponding to an i-AlCuFe QC. A 10 fold symmetry axis around the origin can be clearly appreciated along with a series of nested pentagons, two of which are highlighted at the upper right corner (Courtesy of J. Reyes-Gasga).

for the d-Al₇₀Cu₁₅Co₁₅ QC diffraction pattern displayed in Fig. 1.7 are listed in the second column of Table 1.5 and they clearly follow a non-periodic series, obeying the Fibonacci-like recursion rule $d_{n+1} = d_n + d_{n-1}$. Also, we can appreciate a close correspondence between the listed d_n/d_1 values and successive powers ($n > 1$) of the golden mean $\tau = (\sqrt{5} + 1)/2 \simeq 1.62$ (rounded to the second decimal place), as it is seen in the fourth and fifth columns of Table 1.5. In the third column we list the rational approximants to the decimal expressions given in the fourth column. As we see, all of them share the same denominator, while the numerators are given by the Fibonacci series $\{5, 8, 13, 21\}$, in agreement with the asymptotic limit prescribed by Eq. (1.5), and their natural extensions, as described in Sec. 1.1.2. Finally, taking into account the relation between τ^n powers and Fibonacci numbers given by Eq. (1.3), the corresponding diffraction peaks can be labeled in terms of Fibonacci couples, as it is indicated in the last column of Table 1.5.

TABLE 1.5 The main reflections in the diffraction pattern shown in Fig. 1.7 along a radial direction can be labeled according to a power series related to the golden mean.

n	d_n (mm)	d_n/d_1	d_n/d_1	τ^{n-1}	$ \Delta $	(F_n, F_{n-1})
1	6.0	1	1.00	1.00	—	—
2	10.0	5/3	1.66	1.62	0.04	(1, 0)
3	16.0	8/3	2.66	2.62	0.04	(1, 1)
4	26.0	13/3	4.33	4.24	0.09	(2, 1)
5	42.0	21/3	7.00	6.85	0.15	(3, 2)

(b) Proceeding in a completely analogous way with the diffraction pattern displayed in Fig. 1.27 we obtain the data listed in Table 1.6 for an i-AlCuFe QC.

TABLE 1.6 The main reflection spots along a radial direction in the diffraction pattern shown in Fig. 1.27 can be labeled according to a power series related to the golden mean.

n	d_n (mm)	d_n/d_1	d_n/d_1	τ^{n-1}	$ \Delta $	(F_n, F_{n-1})
1	9.25	1	1.00	1.00	—	—
2	14.50	58/37	1.57	1.62	0.05	(1, 0)
3	23.75	95/37	2.57	2.62	0.05	(1, 1)
4	38.75	155/37	4.19	4.24	0.05	(2, 1)
5	63.50	254/37	6.86	6.85	0.01	(3, 2)

By comparing the results presented in Tables 1.2, 1.5, and 1.6, we realize that the main features displayed by the electron diffraction patterns of decagonal and icosahedral QCs, measured along the 5- and 10-fold axes, respectively, are very similar. In particular, the scale factor is given by the golden mean in both cases (within the experimental accuracy). Nevertheless, by inspecting the data listed in the sixth column of Tables 1.2, 1.5, and 1.6, we note that the i-AlCuFe sample displays a higher structural quality than the i-Al₈₆Mn₁₄ sample, measured in terms of the smaller $|\Delta|$ values.

1.3 *Determine the characteristic inflation scale factor of a dodecagonal QC making use of the Bragg reflections arranged along the 15° radial lines in the diffraction pattern shown in Fig. 1.28.*

The main peaks along the 15° radial lines are labelled F, C, D outwards from the center. The F and D peaks average distances, measured along each of the twelve radial lines, read $d_F = 11.7 \pm 0.2$ mm, and $d_D = 44.1 \pm 0.3$ mm, respectively. Thus, we get the ratio

$$\frac{d_D}{d_F} = \frac{44.1}{11.7} = \frac{49}{13},$$

that is, $\frac{d_D}{d_F} \simeq 3.77 \pm 0.09$, which is reasonably close to the exact value $2 + \sqrt{3} = 3.73205081 \simeq 3.73$, within the experimental accuracy.

1.4 (a) *Making use of the relation $\tau = 2 \cos \frac{\pi}{5}$ deduce the ratio between the diagonal and the side of a regular pentagon (Fig. 1.29). Compare this exact result with the experimental value obtained from the diffraction pattern shown in Fig. 1.7b. (b) Determine the scale factor between the large and small pentagons in the Pythagorean pentagram highlighted in Fig. 1.7.*

(a) Making use of the cosine theorem we have $d^2 = 2a^2 - 2a^2 \cos \varphi = 2a^2(1 - \cos \varphi) = 4a^2 \sin^2 \frac{\varphi}{2} = 4a^2 \sin^2 \left(\frac{3\pi}{10} \right)$, where we have used the value $\varphi = \frac{3\pi}{5}$ for the angles of a regular pentagon. Thus, we get the ratio

$$\frac{d}{a} = 2 \sin \left(\frac{3\pi}{10} \right) = 2 \sin \left(\frac{\pi}{2} - \frac{\pi}{5} \right) = 2 \cos \frac{\pi}{5} = \tau.$$

On the other hand, measuring the length of the five sides and diagonals of the large pentagon highlighted in Fig. 1.7 we obtain the average values $a = 16.2 \pm 0.2$ mm and $d = 26.1 \pm 0.2$ mm, respectively, which yields the ratio $d/a = 1.61 \pm 0.03$, a value which agrees well with that of the golden mean within the experimental accuracy.

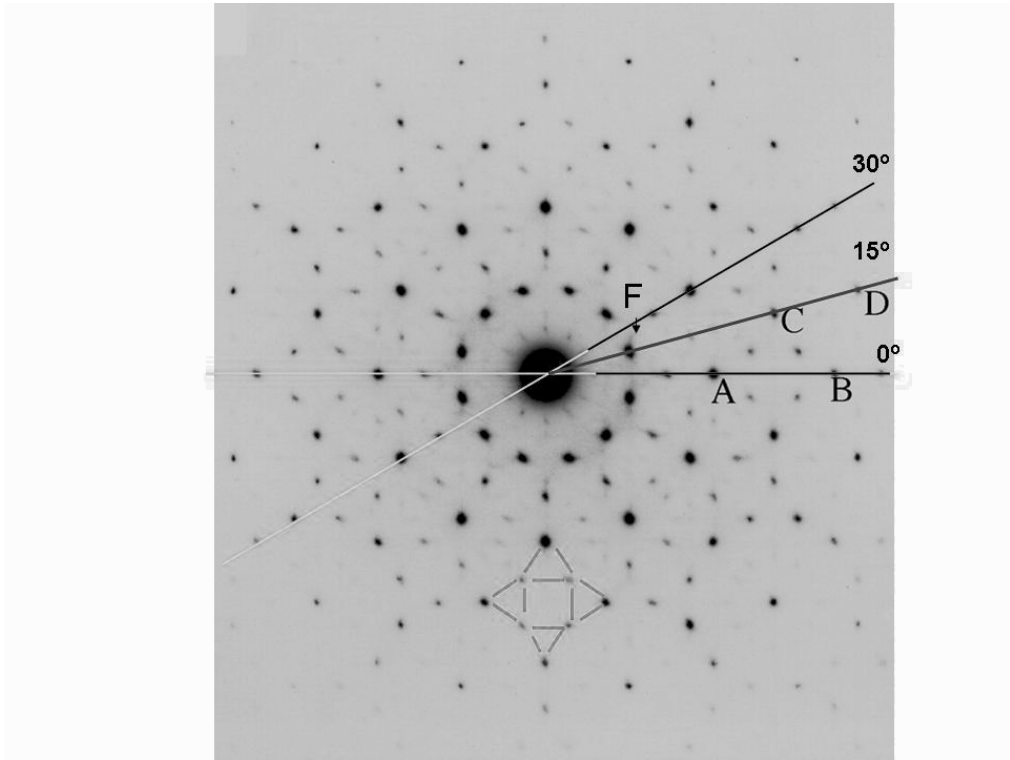


Figure 1.28 Electron diffraction pattern corresponding to a MnCrNiSi dodecagonal quasicrystal taken along the 12-fold axis. Characteristic motives (one of them highlighted at the bottom) based on squares and equilateral triangles sharing a common edge can be readily appreciated. (Adapted from *Dodecagonal quasicrystal in Mn-based quaternary alloys containing Cr, Ni and Si*, by Iwami S, and Ishimasa T, *Philosophical Magazine Letters*, 85, 229–236, 2005. Reprinted by permission of Taylor & Francis Ltd, <http://www.tandfonline.com>).

(b) Measuring the length of the five sides of the first generation small pentagon drawn at the left bottom corner of Fig. 1.7 we obtain the average value $a' = 6.4 \pm 0.1$ mm, so that we obtain the scale factor $a/a' = 2.53 \pm 0.07 \simeq \tau^2$, within the experimental accuracy (see Table 1.5). For the sake of illustration, we have also measured the average side length of the second generation smaller pentagon encircled in Fig. 1.7, obtaining $a'' \simeq 2.5 \pm 0.2$ mm, which leads to the second order scale factor $a/a'' = 6.5 \pm 0.6$, which falls somewhat short from the expected value $\tau^4 \approx 6.85$ (see Table 1.5).

1.5 Following the procedure described in Section 1.1.2 study the relative distances to the center along a radial direction of the main diffraction peaks of the icosahedrite natural QC shown in Fig. 1.15. Compare the obtained results with those reported in Tables 1.2 and 1.6.

Proceeding in a way completely analogous to that described in the **Exercise 1.2a** above, we obtain the data listed in Table 1.7. By comparing the results listed in Tables 1.2, 1.7, and 1.6, we conclude that the natural QC sample (the mineral icosahedrite) displays a

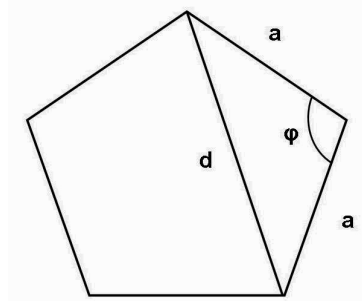


Figure 1.29 Geometrical elements of a regular pentagon.

TABLE 1.7 The main reflection spots along a radial direction in the diffraction pattern shown in Fig. 1.15 can be labeled according to a power series related to the golden mean.

n	d_n (mm)	d_n/d_1	d_n/d_1	τ^{n-1}	$ \Delta $	(F_n, F_{n-1})
1	4.25	1	1.00	1.00	—	—
2	6.75	27/17	1.59	1.62	0.03	(1, 0)
3	11.00	44/17	2.59	2.62	0.03	(1, 1)
4	18.00	72/17	4.23	4.24	0.01	(2, 1)
5	29.00	116/17	6.82	6.85	0.03	(3, 2)
6	46.75	187/17	11.00	11.09	0.09	(5, 3)

significantly higher degree of structural quality than the i-AlMn QC found by Schechtman or typical i-AlCuFe QCs produced in the laboratory samples, as measured in terms of: (1) its smaller $|\Delta|$ values, and (2) the fact that we can observe Bragg reflections corresponding to the τ^5 terms in its electron diffraction pattern. In addition, the more intense Bragg peaks display no discernible evidence of phason strain (note the remarkably isotropic shape of the peaks, as compared with those shown in Fig. 1.27, for instance) [67].

1.6 Determine the global or discrete nature of the scale transformation symmetry acting on the spiral lattice given by the relationships $r_n = a\sqrt{n}$ and $\varphi_n = n\alpha$, where a and α are positive real numbers.

By combining both expressions we get $r_n = a\sqrt{\frac{\varphi_n}{\alpha}} \rightarrow \varphi_n = \frac{\alpha}{a^2}r_n^2$. The action of the scale transformation operator is given by

$$S_\lambda \varphi_n(r_n) = \frac{\alpha}{a^2}(\lambda r_n)^2 = \lambda^2 \varphi_n(r_n),$$

so that we obtain for the discrete spiral lattice the same scale factor $\mu = \lambda^2$ we obtained for the related (Fermat) spiral curve. In this way, we have a discrete lattice which exhibits a global scale invariance symmetry.

1.7 Making use off Eqs. (1.13) and (1.14) prove the scale invariance relationship $S_\lambda \tilde{\mathbf{a}} = \tau \tilde{\mathbf{a}}$, involving the icosahedral lattice vectors $\{\mathbf{a}_i\}$ in 6D.

$$\begin{aligned}
\mathbf{S}_\tau \tilde{\mathbf{a}} &= \frac{1}{2} \begin{pmatrix} 1 & 1 & 1 & 1 & 1 & 1 \\ 1 & 1 & 1 & -1 & -1 & 1 \\ 1 & 1 & 1 & 1 & -1 & -1 \\ 1 & -1 & 1 & 1 & 1 & -1 \\ 1 & -1 & -1 & 1 & 1 & 1 \\ 1 & 1 & -1 & -1 & 1 & 1 \end{pmatrix} \begin{pmatrix} \mathbf{a}_1 \\ \mathbf{a}_2 \\ \mathbf{a}_3 \\ \mathbf{a}_4 \\ \mathbf{a}_5 \\ \mathbf{a}_6 \end{pmatrix} \\
&= \frac{1}{2} \begin{pmatrix} 1 & 1 & 1 & 1 & 1 & 1 \\ 1 & 1 & 1 & -1 & -1 & 1 \\ 1 & 1 & 1 & 1 & -1 & -1 \\ 1 & -1 & 1 & 1 & 1 & -1 \\ 1 & -1 & -1 & 1 & 1 & 1 \\ 1 & 1 & -1 & -1 & 1 & 1 \end{pmatrix} \begin{pmatrix} 1 & 0 & \tau \\ \tau & 1 & 0 \\ 0 & \tau & 1 \\ -1 & 0 & \tau \\ 0 & -\tau & 1 \\ \tau & -1 & 0 \end{pmatrix} \begin{pmatrix} \mathbf{e}_1 \\ \mathbf{e}_2 \\ \mathbf{e}_3 \end{pmatrix} \\
&= \begin{pmatrix} \tau & 0 & \tau+1 \\ \tau+1 & \tau & 0 \\ 0 & \tau+1 & \tau \\ -\tau & 0 & \tau+1 \\ 0 & -\tau-1 & \tau \\ \tau+1 & -\tau & 0 \end{pmatrix} \begin{pmatrix} \mathbf{e}_1 \\ \mathbf{e}_2 \\ \mathbf{e}_3 \end{pmatrix} = \tau \tilde{\mathbf{B}} \tilde{\mathbf{e}} = \tau \tilde{\mathbf{a}}.
\end{aligned}$$

where we have used the relation $1 + \tau = \tau^2$.

1.8 Show that the function $f(x) = \sin(x) + \cos(\sqrt{2}x)$ satisfies the condition $|f(x + P_n) - f(x)| < \varepsilon$, $\forall x \in \mathbb{R}$, for a countable set of almost periods P_n , and estimate their values.

Let $P_n \equiv nP$, where $n \in \mathbb{Z}$, and $P \in \mathbb{R}$, such that $\sin P_n = 0$. Then,

$$\begin{aligned}
|f(x + P_n) - f(x)| &= \left| \sin(x + P_n) + \cos(\sqrt{2}x + \sqrt{2}P_n) - \sin(x) - \cos(\sqrt{2}x) \right| \\
&= \left| \cos(\sqrt{2}x + \sqrt{2}P_n) - \cos(\sqrt{2}x) \right| \\
&= \left| -2 \sin\left(\frac{\sqrt{2}}{2}P_n\right) \sin\left(\frac{\sqrt{2}}{2}(2x + P_n)\right) \right| \leq 2 \left| \sin\left(\frac{\sqrt{2}}{2}nP\right) \right|.
\end{aligned}$$

Because $\sqrt{2}$ is irrational, the sequence $\sqrt{2}nP/2 \pmod{1}$ is dense in the interval $[0, 1)$. It is thus clear that for any $\varepsilon > 0$, there are infinitely many integers $n \in \mathbb{Z}$, so that the relationship $|f(x + P_n) - f(x)| < \varepsilon$ holds for all $x \in \mathbb{R}$ (qed).

In order to estimate the possible values for the almost periods P_n we note that $|\sin(x)| \leq |x|$ on $\mathbb{R}$, so that we can write $\varepsilon = 2 \left| \sin\left(\frac{\sqrt{2}}{2}P_n\right) \right| \leq \sqrt{2}P_n$. Now, $\sqrt{2}$ can be approximated by the members of the series

$$\left\{ 1, \frac{3}{2}, \frac{7}{5}, \frac{17}{12}, \frac{41}{29}, \frac{99}{70}, \frac{239}{169}, \dots \right\},$$

which are of the form $\frac{a_n}{b_n}$, with $a_n = a_{n-1} + 2b_{n-1}$ and $b_n = a_{n-1} + b_{n-1}$, for $n \geq 1$, with $a_0 = b_0 = 1$. Thus, for a given n value we have $P_n \simeq \frac{b_n}{a_n} \varepsilon$ [32].

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