Quasicrystals

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Abstract
This article aims at a simple introduction to the structure, properties and
usefulness of quasicrystals. Since space is limited, the reader will essentially
find a selection of references that may be used to obtain better insight into this
rather new part of materials science.

1. Introduction

For sure, D Shechtman was not taking a nap on 8 April, 1982 afternoon. We know from his
logbook that he immediately noticed that the electron diffraction pattern he observed for the
first time on this very afternoon was a monster [1]. On the one hand, the pattern showed
sharply defined spots. Hence, the material that the electron beam had passed through was a
crystal with a substantial degree of order. On the other hand, it could not be a crystal because
a five-fold point group symmetry was obvious in this pattern. As everybody used to know
then from theoretical crystallography, crystals can exhibit only two-, three-, four- and six-fold
invariance by rotation, and accordingly in their diffraction patterns. This new structure, now
commonly accepted in materials science and solid-state physics, is still not fully solved and it
took about a decade to rationalize the discovery of Shechtman by reference to high dimensional
spaces [2]. Such a material belongs to a series of aperiodic compounds, but is more widely
known as a quasicrystal because the positions of the atoms may be related to quasiperiodic
density functions [3].

Quasiperiodic functions form a most interesting class of mathematical functions, which
may be produced by projecting a high-dimensional periodic function onto a subspace of
lower dimension, appropriately oriented with respect to the high-dim space. Another one-
dimensional example of a quasi-periodic function is given by the sequence of characters
generated by a substitution algorithm: B replaces A, AB replaces B. Starting with AB, and
after three substitution steps, one has BABABBAB. After an infinite number of substitutions,
the chain is necessarily aperiodic, as the reader may like to check. The transport properties
of aperiodic chains of atomic sites mapped onto this kind of sequence were studied in a
systematic way [4]. It was shown that under such circumstances, the electronic states are
no longer extended as in a periodic solid nor localized as in a disordered system. Instead, a
specific kind of states, dubbed critical, was demonstrated to take place. This result lies at the

core of our current understanding of the quite unique behaviour of the physical properties of quasicrystals [5]. Indeed, although formed of 'good' metals like aluminium, copper and iron, or aluminium-palladium and manganese, quasicrystals exhibit conduction properties rather reminiscent of those of semiconductors [6]. Hence, they may be used for various technological purposes [7].

The aim of this article is to introduce the reader to a number of results gained in this new area of materials science since the discovery of Shechtman *et al* [8]. It is also aimed at listing a few of the basic textbooks that may help to gain a better insight into the subject [5, 9, 10].

2. Metallurgy

Within a year after the first report on the existence of aperiodic order in condensed matter, new kinds of quasicrystals were uncovered. Whereas the very first quasicrystal, called the icosahedral compound, is a genuine three-dimensional aperiodic solid [8], two-dimensional quasicrystals do also exist. They consist of periodic stackings along one rotational symmetry axis of puckered, aperiodic layers of atoms. Hence, decagonal quasicrystals, with a ten-fold rotational axis [11], or eight-fold [12] or 12-fold [13] quasicrystals are known. Even one-dimensional quasicrystals, i.e. quasicrystals that exhibit one aperiodic direction, the other two directions being periodic, were discovered [14]. These icosahedral and decagonal quasicrystals were found in binary mixtures of aluminium or titanium alloyed with a transition metal. They proved metastable and returned to normal crystalline state upon heating.

A couple of years after the first publications on quasicrystals, some ternary quasicrystals turned out to be stable and to belong to equilibrium phase diagrams [15]. These quasicrystals may be prepared by conventional metallurgy techniques (melting followed by slow casting) and resist thermal treatments without transforming into normal crystals when annealed in vacuum or under an inert atmosphere [16]. Most of these stable quasicrystals are constituted of aluminium mixed with transition metals like Cu and Fe (or Ni and Co) or normal metals like Mg and Zn. Hence, it happens that quasicrystals are, to a large extent, low-cost materials, easy to produce in large quantities and adaptable to a number of industrial preparation processes (for instance, melting followed by gas atomization and plasma spray) [10, 17]. Stable quasicrystals were also discovered in Ti-based alloys [18] and in the rare earth–Mg–Zn systems [19]. For some time, it was assumed that the stability of quasicrystals imposes one to consider only ternary alloys. This is not true anymore, for a binary quasicrystal was discovered recently in Cd–Ca and Cd–Yb [20].

Of course, like most metals or intermetallic compounds, quasicrystals oxidize when exposed to air or to oxygen [21–23]. The oxide skin that forms under such conditions is thin (roughly below 5 nm), but essentially passivating up to a few hundred degrees Celsius so that oxidation soon stops [24]. Furthermore, the chemical reactivity of quasicrystals in most electrolytes, except strong alkaline solutions, is pretty good and comparable to that of metallic alloys like austenitic steels [25].

3. Crystallography

The discovery by Shechtman *et al* [8] of an unexpected diffraction pattern showing simultaneously sharp diffraction spots and ten-fold symmetry has produced some criticism from the world of conventional crystallography [26]. A few authors, especially Pauling, Nobel Prize winner in Chemistry, rejected the idea of a genuine non-crystalline structure and challenged the occurrence of a new type of order in condensed matter by referring to
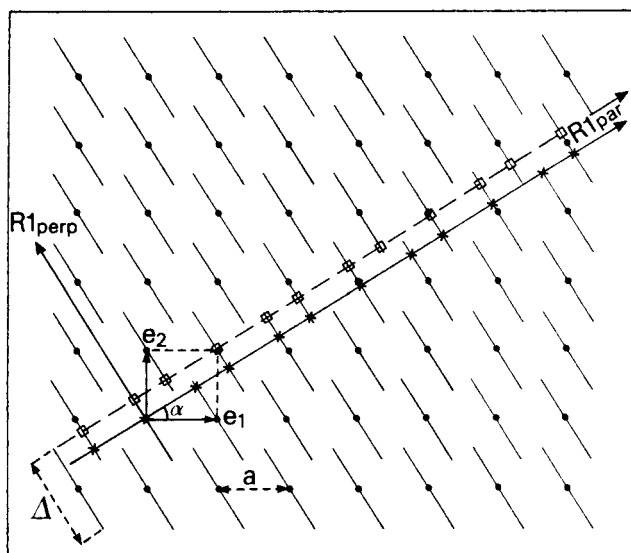
Figure 1. Toy-model of the hyperspace image of a quasicrystal. Here a one-dimensional aperiodic chain of atomic sites is generated along a one-dimensional space denoted $R1_{par}$. Its two-dimensional periodic image is shown as a decorated square lattice (of lattice parameter a). The atomic surfaces, represented by thick segments of length Δ, are arranged in a complementary space, denoted $R1_{perp}$. They are designed in such a way that their intersections with $R1_{par}$ reduce to points, the atomic sites of interest. The chain may become periodic if the space $R1_{par}$ is tilted in such a way that it goes through more than one lattice node. This corresponds to a rational orientation α of $R1_{par}$ with respect to the high-dimensional square lattice. Conversely, an aperiodic sequence of sites is given by an irrational orientation. Movements of $R1_{par}$ parallel to itself generate atomic jumps as is easily observed on the figure when the intersection of $R1_{par}$ with an atomic surface comes close to its extremity. This kind of excitation, specific to quasicrystals, may also occur under the effect of a localized distortion of $R1_{par}$.

micro-twinning artifacts [27]. The first authors, however, who were right on track from the very beginning, could not unambiguously demonstrate the existence of genuine aperiodic icosahedral quasicrystals until the discovery of facetted, stable, mono-domain icosahedral samples [28]. Even more important in this respect was the existence, established systematically in the years following the discovery, of approximant crystals [29]. Such crystals of similar compositions and crystalline structures as the parent quasicrystal are distinguishable from the parent in reciprocal space and exhibit very large, if not giant, unit cells. Hence, the quasicrystal appears to be the limit to infinite size of the unit cell of these approximants, which in turn proves the existence—within the accuracy of our instruments—of true quasicrystals. A tremendous research effort was generated in order to put this knowledge on a firm theoretical basis. Following the original frame worked out by de Wolf *et al* [30] for the crystallography of incommensurate structures, it was demonstrated nearly simultaneously in France, Russia and the United States that quasicrystal structures may be referred to as high-dimensional periodic lattices by cut and projection [31–33]. The atomic positions in the physical, three-dimensional space of the quasicrystal are then generated by a cut of a periodic lattice of higher dimension, the nodes of which are decorated by so-called atomic surfaces (see figure 1). A complementary space, which carries those atomic surfaces, is defined in such a way that its intersection with physical space reduces to simple points where the atoms are located. The high-dimension space results from the combination of physical and complementary spaces. By definition

of Fourier transformation, the physical reciprocal space of the quasicrystal corresponds to the projection onto it of the Fourier transform of the high-dim periodic lattice convoluted by the Fourier transform of the atomic surface. This scheme enables us to generate either three-dimensional icosahedral quasicrystals by cut of a six-dimensional hypercubic lattice, or two-dimensional quasicrystals, by stacking along a periodic direction aperiodic layers of atoms obtained by a cut of four-dim periodic lattices along different directions (and with different atomic surfaces). Accordingly, atomic layers may exhibit eight-fold, ten-fold or 12-fold rotational symmetry, all of forbidden type in conventional crystallography. This new high-dimensional crystallography generalizes the conventional crystallography known before the discovery of quasicrystals [2, 5, 9]. It encompasses with no doubt the most important output that this discovery has brought to modern solid-state physics. Application to real quasicrystals in order to decipher atomic positions from diffraction patterns was indeed rather successful [29, 34–36], although some limitations of the technique do not allow as yet a sufficient degree of accuracy on every atomic site. These limitations are too technical for the purpose of the present review, see [29, 37].

The intersection of physical space with the atomic surfaces defines one specific sequence of atomic sites. This sequence depends naturally on the specific position of physical space within the high-dimensional lattice. Another position along the components of complementary real space (with the same angular orientation) would define another sequence, yet thermodynamically equivalent since all positions are arbitrary and hence equivalent. However, small fluctuations of those positions or, more importantly, small localized distortions of the cutting physical space may occur under the effect of thermal excitations at finite temperature. In each collection of atomic sites generated by this algorithm, there exist a number of sites, which are produced by cutting very close to an edge of an atomic surface. Those sites may disappear with a fluctuation of the cut space, but must re-appear close by in order to preserve the fixed atomic density. Therefore, the size, symmetry and respective positions of the atomic surfaces are defined accordingly [38] and small fluctuations of the cut space are associated with small atomic jumps called phason flips that preserve the overall atomic density under the effect of thermodynamic fluctuations above zero absolute temperature.

This notion of phasons flips appears as a natural by-product of generalized crystallography, but must also correspond to real excitations in physical space. On top of the experimental detection of such localized atom jumps, both at high [39] and at low [40] temperatures, it turns out that this concept is at the heart of our current interpretation of the physical and mechanical properties of quasicrystals. Computer simulations [41] and theory [42] tend to associate such phason flips with an enhancement of the atomic diffusion in the medium temperature range. Presumably, the plastic behaviour of quasicrystals at the approach of the melting temperature is also most related to the increasing density of phason flips that take place above typically 550 °C [43]. In addition, a static phason strain field may be associated with the disorientation of the cut space within the high-dimensional reference lattice, thus offering an opportunity to describe the phase transitions often observed between a quasicrystal and its approximant crystals [29]. From hydrodynamic theory [44], such a transition is triggered by a phason mode, evidence of which was pointed out in the anisotropy and momentum transfer dependence of the diffuse intensity measured at the bottom of strong diffraction peaks of icosahedral mono-domain samples [45].

4. Properties and applications

An exhaustive list of all properties studied so far in quasicrystals would need a full textbook (see e.g. [10, 11]) and would go beyond the aim of the present review. Therefore, I restrict myself

to a few of those properties that may find a technological application [7]. Despite the fact that quasicrystals may be taken at first glance as metallic alloys owing to the nature of their constituents, they do not behave like conventional intermetallic compounds. They exhibit fascinating and, as it turns out, useful properties as well. Academic evidence of this fact is the electrical resistivity. At low temperature (4 K), the electrical resistivity is about three orders of magnitude larger than that of the individual metallic constituents. Upon heating, it decreases but is enhanced if the structural perfection of the quasicrystal is improved. These trends are opposite to the behaviour of good metals like aluminium and resemble more that of a semiconductor [6]. In some icosahedral compounds like Al–Pd–Re, evidence exists that a metal–insulator transition takes place close to zero absolute temperature [6, 46]. The absence of any Drude peak on the optical conductivity at low frequencies and the appreciable absorption of infra-red and visible light are also clear indications that electronic transport in quasicrystals is specific. This holds true as well for the phonon density of states which shows only well defined acoustic modes of energy (frequency) below 2 THz [9]. As a consequence, heat conduction is very much reduced and compares well to that of doped zirconia, even up to the melting temperature at typically 1200 K [47]. Keeping in mind[1] that the expansion coefficient of quasicrystals is found in the range 14.0–19.0×10^{-6} K^{-1}, and that these materials soften when their temperature exceeds about 600 °C, are strong points in support of the use of quasicrystals for the manufacture of thermal barriers adapted to heat insulation in car engines, vapour turbines, some parts of aircraft compressors or cryogenic devices [48].

In fact, quasicrystals are brittle materials at temperatures below about 500 °C, with toughness constants exceeding only marginally 1 MPa m$^{1/2}$ when the grain size is very small. By contrast, they become essentially ductile when the temperature rises above 500 °C, with a marked plastic regime and no work hardening [43]. This specific behaviour is related to the nucleation and dynamics of dislocations, like in normal crystals [49]. However, both experimental and computer simulation studies indicate that the mobility of dislocations is associated with a climb mechanism that involves a recovery via phason flips of the icosahedral lattice, hence explaining the absence of work hardening [50]. Simultaneously, the glide of a dislocation in a quasicrystal encounters energy barriers when passing through well defined and stable clusters of atoms [51] which also contribute to the observed stress–strain curves [52].

All quasicrystals based on aluminium show a pronounced pseudo-gap in the Al partial densities of states evaluated at the position of the Fermi energy [53]. The same conclusion is reached for closely related intermetallics, especially approximants of high order. The pseudo-gap is therefore not a signature of quasicrystalline order, although its depth and width are maximized in the true quasicrystals with the best lattice perfection. Not surprisingly, the depth of the pseudo-gap correlates with the transport properties and the stability of the quasicrystal [54]. In Al–transition metal quasicrystals, the existence of a pronounced pseudo-gap in the Al 3p and 3s,d partial densities of states is to a large extent responsible for the enhanced stability of the quasicrystal via a Hume–Rothery mechanism [55]. The existence of the pseudo-gap is not restricted to the bulk lattice, but persists up to the very surface of well prepared, clean icosahedral and approximant samples [56].

Wetting with water, hence adhesion of a polar liquid, on the surface of quasicrystals is also reduced in comparison with metallic or oxide surfaces. This behaviour cannot be anticipated from the current knowledge of the constituent metals and more significantly from that of the oxide always present at the surface of quasicrystals in ambient conditions. As it appears, this behaviour is related to the specific electronic structure of quasicrystals which determines the

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[1] In comparison, pure aluminium shows a thermal expansion coefficient of 24×10^{-6} K^{-1} at room temperature. Its electronic resistivity is well below 2 $\mu\Omega$ cm, depending on sample purity. In single crystals of very high quality, it is as small as 0.3 $\mu\Omega$ cm at 4 K. The thermal conductivity is typically equal to 200 W m^{-1} K^{-1} at 300 K.

electronic interaction—across the oxide film—of the H_2O dipole with the partially localized cloud of conduction electrons in the quasicrystal. In fact, the reversible adhesion energy of water correlates with the electronic density at the Fermi edge [57]. A direct application of this property, namely the use of quasicrystalline coatings for the production of scratch resistant films offering a lowered adhesion to food or to some polymers, is already on the market [58].

In the case of perfect quasicrystals at least, strong evidence exists that the reduced friction forces which they oppose to solid riders is in part due to the absence (or decrease) of chemical bonding at the common interface. Furthermore, the unfavourable matching of atomic lattices of a crystalline indenter and of a quasicrystal, together with its great hardness of the order of 0.8–1.1 GPa, reduce the friction coefficient μ. For instance, figures as small as $\mu = 0.04$ with respect to diamond or $\mu = 0.1$ with respect to sintered WC–Co balls were achieved [59]. This opens the way for many tribological applications in mechanical devices, although the brittleness intrinsic to perfect quasicrystals limits this view to films supported by a ductile substrate or to quasicrystal sinters tailored like technical ceramics.

Many other applications of quasicrystals were foreseen in recent years in fields as different as catalysis [60], thermo-power generation [61], hydrogen storage [18], solar light absorption [62], reinforcement of metal–matrix [63] and polymer-matrix [64] composites and, last but not least, precipitation hardening of maraging steels [65] and Al-based light alloys [66] incorporating nanometre sized quasicrystalline particles. These latter products are now commercially available, especially in electric shavers comprising different parts made of a maraging steel reinforced with nano-icosahedral particles. To the best of my knowledge, this is actually the second successful application of quasicrystal on the market.

There is no need to emphasize here the fact that quasicrystals were disclosed in the literature in 1984, only 16 years ago, and that they are therefore new materials, not as mature by far as most materials in use in today's technology. Many unknowns still remain, including major questions regarding their formation and thermodynamic stability. Although potentially important for a range of applications—in short, related to energy savings—processing of quasicrystals in view of their production and use on an industrial scale is still very much a matter of future research and a venture that only a few companies have undertaken [67].

5. Beyond the science of quasicrystals

Aside from studies on materials [10] and beyond their tight relationship to mathematics [2], the usefulness of quasicrystals is manifest because the concepts born in this field will find more applications in quite different scientific domains. To argue on this standpoint, I will give only a few examples taken from works published in solid and fluid mechanics and in low-temperature physics.

A major problem in mechanics, and even more so in civil engineering and architecture, is that solid materials are not able to sustain constraints equally well in all directions of space. Wood for instance is an outstanding material for buildings provided forces act in the right directions. Concrete cannot resist stresses except in uniaxial compression. Many attempts have been made to solve these problems and composite materials offer a solution, the efficiency of which depends on the symmetry of the arrangement of the inner filling material, yet being of limited value because of the simple architectures used in most cases. An icosahedral array of filling fibres would naturally optimize the isotropy of the partition of internal stresses, but it was long believed that a filling of maximal density—as required to maximize the efficiency of the composite material—could not be achieved on practical grounds. Duneau and Audier proved that this is not true [68]. They demonstrated that high-dim crystallography techniques must be used to design a composite filling with maximal density. Macroscopic samples were

fabricated and tested on mechanical benches, as well as computer simulations using finite element codes, used to explore different filling configurations. From these studies it was found that the response of these composites to mechanical stresses is indeed quite isotropic and therefore these findings can be used in further developments in mechanical sciences. The work by Duneau and Audier opens a new way of filling architectural space in three dimensions, yet with a better response to external constraints, which architects and designers will hopefully exploit quite soon. Furthermore, this type of composite, when prepared from an optically transparent medium, may also exhibit photonic pseudo-gaps of great interest from the viewpoints of academic understanding and technological applications.

High-dimensional crystallography was recently used to explain how Faraday waves form at the surface of a vibrating liquid and how this may result in dodecagonal patterns [69]. The existence of this phenomenon is rather ancient since Faraday [70] discovered it nearly two centuries ago in an open vessel containing a fluid that he subjected to vertical sinusoidal vibrations, thus modulating periodically the effective gravity. A more recent experiment was performed in the 1990s, which using sophisticated imaging systems allowed the detection of the formation of stripes, squares, hexagons and dodecagons at the surface of the liquid when the vertical excitation wave was composed of a linear combination of two frequencies. Depending on the respective amplitude and phase shift of the two waves, the resulting pattern thus exhibited two-, four-, six- or 12-fold symmetry, but eight- and ten-fold patterns were not observed. This phenomenon had not received a satisfactory interpretation until the work by Lifshitz and Petrich [69]. These authors used a model equation for the movement of the fluid surface across its plane at rest, which is a standard in the mechanics of fluids to show supercritical instabilities. I do not need to reproduce this equation here since we are more interested in the method of thinking used by the authors in relation to quasicrystals rather than in the detailed results. This equation contains a positive-definite gradient term which selects only those waves which are specified by a wave vector close to a given critical value, e.g. $k_c = 1$. A supplementary cubic term is added to account for the coupling of triad interactions, which also minimize the free energy of the system and form hexagonal patterns. Then, guided by their knowledge of quasiperiodic patterns, the authors modified the standard equation and introduced a double positive-definite gradient term able to select only modes which are near one of two critical values, $k_c = 1$ or q. They defined q as the magnitude $q = 2\cos(\pi/12)$ of the vector sum of two unit vectors separated by an angle of $30°$. This parametric equation depends only on two wave vectors of magnitude 1 and q and two control parameters, which monitor the linear and quadric displacements. Searching for the minima of the free energy results in a phase diagram, which reproduces adequately the experimental evidence available so far. The region where dodecagonal patterns are stable is well defined and computer simulations performed with parameters chosen in this region come out in excellent agreement with the experimental data. Patterns with eight- or ten-fold symmetry cannot form under these conditions, also in accordance with observations.

In low-temperature physics, the use of a combination of laser beams converging towards a small region, into which they interfere and hence form a trap for resonating atoms, is the key that has opened the way to extremely low temperatures in the range of nanokelvins. The pumping system of coherent light acts on the trapped atoms like ions in solids interact with electrons and provides an effective potential, which slows down the motion of the atoms. In the simplest design of this experiment, the array of laser beams is periodic. Using, for example, six beams directed along the four-fold axes of the cube allows the temperature to be decreased to values so close to absolute zero that Bose condensation effects become manifest. This arrangement may also be chosen as aperiodic, for instance with five beams separated by $72°$ in the plane of the experiment plus eventually two other beams propagating in opposite directions

along the normal to this plane [71]. One relevant result demonstrates that the optical potential shows intensity maxima forming nothing other than a ten-fold lattice in the experimental plane. The efficiency of this trap exceeds that of a periodic optical array by at least 20%. Once more, the concepts derived from high-dimensional crystallography have proved useful outside the field for which they were invented. Many technological applications are foreseen for the cold traps. The most famous one is their use in ultra-precise clocks, able to measure time to better than 10^{-12} s. Therefore, the enhanced efficiency of the aperiodic optical lattices may well carry on one of their most promising breakthroughs.

6. Conclusion

The present review was aimed at assembling a number of key references that the interested reader may use for further understanding the science of quasicrystals. Meanwhile, it was also suggested how this science might trigger new developments in areas no longer linked with materials. Whereas I have commented on some potential applications of quasicrystalline materials, I have deliberately ignored many active subjects for the sake of brevity of the present article. Examples of such topics are surface physics, with the recently discovered new frontier of the complex surface crystallography of aperiodic solids [72], mechanical properties, magnetism, electron localization, corrosion resistance, optical properties, etc as well as the search for finding new quasicrystals in non-metal based systems [73]. The reader is invited to consult the general references listed hereafter for more information on these subjects.

Quasicrystals are already almost two decades old. Nowadays, I have the personal feeling that quasicrystals will go through a period of better understanding, thanks to the progress made on determining their crystalline structures, mostly using advanced synchrotron sources. In contrast, the evidence that new technological developments will show up in the near future is far less obvious, despite the many efforts dedicated to this subject in various countries.

Acknowledgments

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