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Generalized Dynamics of Soft-Matter Quasicrystals

Mathematical Models, Solutions and
Applications

Second Edition

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Mathematical Models, Solutions
and Applications

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Preface to the Second Edition

Due to limited experimental data reported since the publication of the first edition, the study on generalized dynamics of soft-matter quasicrystals has not achieved great progress. In this second edition, we still mainly focus on the mathematical model but provide some applications additional to the first edition.

Nevertheless, there are still some steady progress in the experimental study in the field. The observation of the 10-fold symmetry soft-matter quasicrystal is an example,¹ the importance of which will be beyond that of the first discovered 12-fold one, although the details have not been openly discussed yet. This finding extended the presence of soft-matter quasicrystals in a broader range and would bring fruitful expectations in both experimental and theoretical studies.

Another progress is the experimental and theoretical study on the correlations between the Frank-Kasper phase and soft-matter quasicrystals in giant molecules. This contribution was made by the group of Prof. Stephen Z. D. Cheng and will be introduced in Chap. 2 in the second edition.

In theoretical aspects, we will also introduce the recent progress.

In this circumstance, we strengthen the discussions on possible applications of the theory and the method in new areas, for example, thermodynamic stability, three-dimensional problems, device physics, liquid crystals, general soft matter, etc., which will be introduced in Chaps. 13–16. Of course, the applications are in the preliminary phase.

Thanks to the suggestions from the readers, we paid more attention to supplement recent computational models and simulation results in the second edition to visualize the theoretical formulas. In addition, although errors and mistakes in the first edition have been corrected, there might still be errors and mistakes. The authors are grateful to any criticism from the readers so that we can continuously improve the book.

¹ The 10-fold symmetry quasicrystals in soft matter will be reported by Expanding Quasiperiodicity in Soft Matter: Decagonal Quasicrystals by Hierarchical Packing Frustrations. Authors: Yuchu Liu, Tong Liu, Xiaoyun Yan, Qing-Yun Guo, Huanyu Lei, Zongwu Huang, Rui Zhang, Yu Wang, Jing Wang, Feng Liu, Feng-Gang Bian, E. W. Meijer, Takuzo Aida, Mingjun Huang, Stephen Z. D. Cheng, Proc. Natl. Acad. Sci., in press, 2021.

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The authors are grateful to researcher in metallic materials Prof. Zi-Tong Li, former student Fang Wang from Beijing Institute of Technology for their hard work during the preparation of this second edition.

Due to their important contributions to the second edition, Wenge Yang, Hui Cheng, and Xiao-Hong Sun are invited as co-authors to take charge of the book with me.

Beijing, China
December 2021

Tian-You Fan

Preface to the First Edition

Since 2004, quasicrystals have been discovered in various kinds of soft matters, including liquid crystals, colloids, polymers, and nanoparticles. In particular, 18-fold symmetry quasicrystals in colloids were observed in 2011. More recently the quasicrystals with 12-fold symmetry were also found in surfactants. The formation mechanisms of these kinds of quasicrystals are associated closely with the self-assembly of spherical building blocks by supramolecules, compounds, and block copolymers, and so on, which is quite different from that of the metallic alloy quasicrystals. They can be identified as soft-matter quasicrystals, exhibiting natures of quasicrystals with soft-matter characters. Soft-matter behavior is between solid and simple fluid, while the quasicrystals form in highly ordered structures with crystalline-forbidden symmetry. These features are very complex yet extremely interesting and attractive. Since 2004, soft-matter quasicrystal studies have attracted attention in mathematics, physics, chemistry, and materials science.

So far all observed soft-matter quasicrystals are two-dimensional quasicrystals. It is well known that two-dimensional quasicrystals consist of only two distinct types, one presents 5-, 8-, 10-, and 12-fold symmetries, the other 7-, 9-, 14-, and 18-fold ones according to the group symmetry theory. Therefore, two terminological phrases can be defined such as the first and second kinds of two-dimensional quasicrystals respectively. The two-dimensional solid quasicrystals observed so far only belong to the first kind, while soft-matter quasicrystals discovered up to now belong to both kinds. It is likely other soft-matter quasicrystals beyond 12- and 18-fold symmetries could be found. Therefore, it is quite important to develop a suite of general principles to describe elasticity and fluid dynamics.

It is quite difficult to study these new phases due to the complexity of their structures and limited experimental data including the basic physical constants. It is true for theoretical studies as well. For example, the symmetry groups of soft-matter quasicrystals have yet been thoroughly investigated, and only some preliminary work has been reported so far (the details are not included in the book). In conjunction with this issue, the study on constitutive laws for phason and phonon-phason coupling is still difficult.

Despite only very limited experimental and theoretical studies reported so far, there is great potential to push these directions forward. For example, the soft-matter quasicrystals as a new ordered phase are connected with symmetry breaking, like those discussed in solid quasicrystals. Thus, elementary excitations such as phonon and phason are important issues in the study of quasicrystals based on the Landau phenomenological theory. For soft-matter quasicrystals, furthermore, another elementary excitation—fluid phonon will be considered besides phonon and phason. According to the Landau school, the liquid acoustic wave is a fluid phonon (refer to Lifshitz E M and Pitaevskii L P, *Statistical Physics, Part 2*, Pergamon, Oxford, 1980). This is suitable for describing the liquid effect of soft matter, which can be seen as complex liquids or structured liquids. The elementary excitations—phonon, phason, and fluid phonon and their coupling terms constitute the main feature of these new phases. They will be discussed as a major issue through this book. The fluid phonon was first introduced in the quasicrystals study. Correspondingly, the equation of state should also be introduced in soft-matter quasicrystal study. Combining these two key concepts with the hydrodynamics principle established in solid quasicrystals, the dynamics of soft-matter quasicrystals can be constructed. For solid quasicrystals, there has been tremendous progress over last decades, for example, Lubensky TC, Symmetry, elasticity, and hydrodynamics in quasiperiodic structures, in *Introduction to Quasicrystals*, ed by Jaric MV, Boston: Academic Press, 190-289, 1988; Hu ZC et al, Symmetry groups, physical property tensors, elasticity and dislocations in quasicrystals, *Rep. Prog. Phys.*, 63(1), 1-39, 2000; Fan T Y, *Mathematical Theory of Elasticity of Quasicrystals and Its Applications*, Beijing: Science Press/Heidelberg, Springer-Verlag, 1st edition, 2010, 2nd edition 2016. Based on the development of the theory of solid quasicrystals, we will extend the quantitative analysis into the rich phenomena of soft-matter quasicrystals.

Some applications will be discussed on the distribution, deformation, and motion of soft-matter quasicrystals. The mathematical principles and applications required are briefly reviewed in the first six chapters of this book (for more details, refer to Chaikin J and Lubensky TC, *Principles of Condensed Matter Physics*, New York: Cambridge University Press, 1995). The computational applications on soft-matter quasicrystals are quite preliminary, but they verified partially the mathematical model and explored the distinguished dynamic behavior from solid quasicrystals. In addition, specimens and flow modes adopted in the computation modeling might be intuitive, observable, and can be verified easily.

The author thanks the National Natural Science Foundation of China and Alexander von Humboldt Foundation of Germany for their financial support over the years and Professors U. Messerschmidt from Max-Planck Institut für Mikrostrukturphysik in Halle, H.-R. Trebin from Stuttgart Universitaet in Germany, T. C. Lubensky from the University of Pennsylvania, Stephen Z. D. Cheng from the University of

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December 2016

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**Correction to: Introduction on Elasticity and Hydrodynamics
of Solid Quasicrystals C1**

Notations

\mathbf{r}	Radius vector
D	Domain
S	Boundary of domain
S_u	Boundary part where the displacements are given
S_t	Boundary part where the tractions are given (or S_σ where the applied stresses are given)
ρ	Mass density (g/cm^3)
p	Fluid pressure ($\text{Pa} = \text{N/m}^2$)
\mathbf{u}	Phonon type displacement field (cm)
\mathbf{w}	Phason type displacement field (or second phason displacement field for second kind quasicrystals with 7-, 9-, 14-, 18-fold symmetry) (cm)
\mathbf{V}	Fluid velocity field (or fluid phonon field) (cm/s)
$\varepsilon_{ij} = \frac{1}{2} \left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right)$	Phonon strain tensor
$w_{ij} = \frac{\partial w_i}{\partial x_j}$	Phason strain tensor (or second phason strain tensor for second kind quasicrystals with 7-, 9-, 14-, 18-fold symmetry)
$\dot{\xi}_{ij} = \frac{1}{2} \left(\frac{\partial V_i}{\partial x_j} + \frac{\partial V_j}{\partial x_i} \right)$	Fluid phonon deformation tensor (1/s)
σ_{ij}	Phonon stress tensor (Pa)
H_{ij}	Phason stress tensor (or second phason stress tensor for second kind quasicrystals with 7-, 9-, 14-, 18-fold symmetry) (Pa)
σ'_{ij}	Viscous stress tensor (Pa)
$p_{ij} = -p\delta_{ij} + \sigma'_{ij}$	Fluid phonon stress tensor (Pa)
C_{ijkl}	Phonon elastic coefficient tensor (Pa)
K_{ijkl}	Phason elastic coefficient tensor (or second kind phason elastic coefficient tensor for quasicrystals with 7-, 9-, 14-, 18-fold symmetry) (Pa)
R_{ijkl}	Phonon-phason coupling elastic coefficient tensor (u-w coupling elastic coefficient tensor) (Pa)

η	First viscosity coefficient of fluid ($0.1 \text{ Pa} \cdot \text{s} = \text{Poise}$)
η/ρ	First kinetic viscosity coefficient of fluid (cm^2/s)
ζ	Second viscosity coefficient of fluid ($0.1 \text{ Pa} \cdot \text{s} = \text{Poise}$)
ζ/ρ	Second kinetic viscosity coefficient of fluid (cm^2/s)
Γ_u	Phonon dissipation coefficient ($\text{m}^3 \cdot \text{s}/\text{kg}$)
Γ_w	Phason dissipation coefficient (or second kind phason dissipation coefficient tensor for quasicrystals with 7-, 9-, 14-, 18-fold symmetry) ($\text{m}^3 \cdot \text{s}/\text{kg}$)
\mathbf{v}	First phason type displacement field (only for second kind quasicrystals) (cm)
$v_{ij} = \frac{\partial v_i}{\partial x_j}$	First phason strain tensor (only for second kind quasicrystals)
τ_{ij}	First phason stress tensor (only for second kind quasicrystals) (Pa)
r_{ijkl}	Phonon-first phason coupling elastic coefficient tensor (or u-v coupling elastic coefficient tensor only for second kind quasicrystals) (Pa)
Γ_v	The first kind phason dissipation coefficient of quasicrystals ($\text{m}^3 \cdot \text{s}/\text{kg}$)

Chapter 1

Introduction to Soft Matter



Soft-matter quasicrystals are observed in liquid crystals, colloids, polymers, and surfactants, etc., which brings new family members to soft matter with crystallographic forbidden symmetry. Soft matter is a type of common material, introduced by de Gennes [1] in 1991, including liquid crystals, colloids, polymers, foams, emulsions, surfactants, biomacromolecules, etc. They are neither ideal solid nor simple fluid, but presents characteristics of both solid and fluid, and belongs to an intermediate phase between isotropic fluid and ideal solid macroscopically. Sometimes one calls them anisotropic fluids, structured fluids, or complex fluids [2–5], more exactly speaking, as anisotropic liquids, structured liquids, or complex liquids.

As pointed out by Guo [6], if every atom of a molecule possesses the thermal energy $k_B T$ in an ideal solid, e.g., solid crystal, here k_B being the Boltzmann constant, T the absolute temperature, the thermal energy per unit volume $k_B T/l_0^3$, may characterize an entropy state of the crystal, here $l_0 \sim 0.1$ nm the typical lattice size or interatomic distance. For soft-matter systems, the structure and dynamic properties are related to mesoscopic size $l \sim 10 - 100$ nm (e.g., the size of the long-chain of polymers, or the size of self-assembly structures, etc.). Fluctuation, thermal motion, and self-organization or self-assembly are often induced by entropy with thermal energy per unit volume $k_B T/l^3$. Apparently, at room temperature, the thermal energy per unit volume of soft matter is lower by 6–9 orders of magnitude than that of the ideal crystals. This may explain the softness of soft matter from the perspective of the intra-structure of materials. In contrast, the ideal solid presents very high stiffness. The distinction between soft matter and ideal solid is significant. The thermal energy per unit volume concept may provide a basis by some analogies between soft matter and ideal solid. The other differences between soft matter and conventional materials will be discussed in the following description, but won't be elaborated in detail or in depth here.

For simplicity, we here only consider hydrodynamics, or generalized dynamics, of soft-matter quasicrystals. More strictly speaking, only the fluidity or the flow effect from the perspective of fluid is considered apart from elasticity and interaction between fluidity and elasticity of the matter. The fluidity, elasticity, and their

interaction are the only portions of the behavior of soft matter, which would help us to understand the distribution, deformation, and motion of soft-matter quasicrystals in a microscope. In this case, the micro-scale structures of the matter have not been concerned. Although the mesoscale structures are important for soft matter, it has not been considered in general in our presentation apart from some special exceptions. In this sense, the modeling on hydrodynamics or generalized dynamics of soft matter and soft-matter quasicrystals is a macro-and continuum-medium-study, with low-frequency and long-wavelength characteristics, which have been discussed in solid quasicrystals, and will be extended to the soft-matter case.

Among various kinds of soft-matter systems, liquid crystals are typical and relatively well studied, and their material constants have been reported more in detail. From a macroscopic and continuum point of view, a similar process from liquid crystals can be extended to the study of soft-matter quasicrystals. For example, the generalized Newton's fluid law can approximately be used in some cases, and the generalized Hooke's elasticity law can also be applied, but the deformation in soft-matter quasicrystals is more complex. The deformation of liquid crystals consists of bulk deformation and local curvature variation. For the bulk deformation the conventional generalized Hooke's law still holds, but the deformation induced by curvature variation needs additional terms to be included, which are beyond the discussion in this chapter, and we will return to this issue in Chap. 16 where we discuss the curvature of smectic A liquid crystals. As an intermediate phase between simple fluid and ideal solid, the soft matter presents many behavior differences from those of isotropic liquids and ideal crystals. For example, in ordinary liquid and nematic liquid crystals, there is only one acoustic wave, i.e., longitudinal sound wave. In solid crystals and amorphous solids, there are three acoustic wave speeds under the linear deformation, i.e., $c_1 = \sqrt{\frac{\lambda+2\mu}{\rho}}$ or (or $c_1 = \sqrt{\frac{L+2M}{\rho}}$), $c_2 = c_3 = \sqrt{\frac{\mu}{\rho}}$ (or $c_2 = c_3 = \sqrt{\frac{M}{\rho}}$), as discussed in Chaps. 7–11 of this book. Smectic A liquid crystal has only one nonzero displacement component, the longitudinal shear state in the elastic deformation, so it is often categorized as a one-dimensional crystal. For a pure solid, the acoustic wave speed is $\sim \sqrt{\frac{E}{\rho}}$ where E is the elastic modulus, ρ the mass density; For a pure fluid, the acoustic speed is $\sim \sqrt{\frac{\partial p}{\partial \rho}}$, p the fluid pressure. For smectic A liquid crystals, there are both acoustic wave speeds $\sim \sqrt{\frac{E}{\rho}}$ and $\sim \sqrt{\frac{\partial p}{\partial \rho}}$, where the first speed often depends on the angle between wave vector \mathbf{k} and the normal vector \mathbf{n} of the layer of the smectic A liquid crystal. In general, soft matter behaves differently from simple fluid and ideal solid. Due to complicated nonlinear behavior, the spectra and dispersion relations of soft matter cannot be easily determined, so as the wave speeds. Often one introduces $\sqrt{\frac{\lambda+2\mu}{\rho}}$, $\sqrt{\frac{\mu}{\rho}}$ and $\sqrt{\frac{\partial p}{\partial \rho}}$ (in some cases we denote $\sqrt{\frac{\partial p}{\partial \rho}} = c_4$ for simplicity) to describe wave propagating speeds in the soft matter as a coarse approximation, and the realistic wave speeds in the matter present quite different in magnitude and nature, and so far the relevant mechanism is not yet clear. The successful introduction to the computation can partly reveal these questions.

For liquid crystals, the dynamic viscosity coefficient η (in the unit 1 Poise = 0.1 Pa s) is introduced to describe the fluid effect. Sometimes, the kinetic viscosity coefficient η/ρ also is used, note that the unit is cm^2/s rather than Poise. The elastic modulus E (in the unit $10^8 \text{ erg}/\text{cm}^3 = 10^7 \text{ Pa} = 10 \text{ MPa}$) is used to describe the bulk deformation. The Poisson ratio ν may be negative unlike that for solid. These basic material constants are fundamental and useful for us to deal with the mechanical problems of soft-matter quasicrystals in the late chapters. The viscosity of liquid crystals is quite large (about 100 times that of water), and liquid crystals present a certain degree of elasticity behavior. In general, people do not consider elasticity in simple fluid, and viscosity in ideal solid (at least they are not so important). In the following chapters, we carry out the analysis and computation on distribution, deformation, and motion of soft-matter quasicrystals following the successful experiences from the study of the liquid crystal. Apart from these, some physical constants, e.g., the phonon dissipation coefficient Γ_u and phason dissipation coefficient Γ_w for soft-matter quasicrystals are not available from experimental reports, so we just take relevant values from solid quasicrystals [7] as references.

Another important feature of motion in soft matter is its small Reynolds number Re . According to the definition, $\text{Re} = \frac{\rho U a}{\eta}$, where a represents the characteristic size of the matter or flow field. Because the characteristic velocity U is small and the viscosity coefficient η is large, in general, the Reynolds number is small, i.e., $\text{Re} = 10^{-4} \sim 1$. In this case, the force due to viscosity is much larger due to inertia. Omitting the inertia terms, we can take the Stokes assumption in the equation of motion sometimes, like in the classical fluid dynamics. This simplifies the governing equations which are still very complicated. Not like in classical fluid dynamics where one has obtained quite a lot of approximate analytic solutions, the analytic solution or even approximate analytic solution cannot be derived. Although the governing equations in classical fluid dynamics are complex, they are much simpler than those in generalized dynamics of soft matter. It should be pointed out that the Stokes approximation in a two-dimensional case leads to the famous Stokes paradox—there was no solution. Oseen [8] analyzed the Stokes paradox physically in depth. To overcome the paradox, the Stokes approximation equations must be modified. They should be replaced by Oseen approximation equations and yield reasonable solutions in the two-dimensional case. Further discussion on this issue can be found in Sommerfeld [9], Sleozkin [10], and Kochin et al. [11]. When we discuss the soft matter dynamics, especially the two-dimensional problems, we will get in touch with similar problems, and the Oseen theory provides a useful guideline. Note that the Ref. [10] points out Oseen approximation holds for the cases $\text{Re} < 10$, which is helpful for the study of soft matter.

In addition, the above introduction regarding the soft matter is very limited and preliminary, which only provides the most elementary knowledge for presentation and application in the current chapter. Readers are suggested to refer to monographs [2–5] for a broader understanding of soft matter and related research findings. The generalized dynamics and possible generalized dynamics will be introduced in the subsequent chapters.

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Chapter 2

Discovery of Soft-Matter Quasicrystals and Their Properties



2.1 Experimental Observation of Quasicrystalline Phases in Soft Matter

Quasicrystals have long-range orientational order but no translational symmetry. As a consequence, sharp diffraction spots can occur but are unable to be described by 230 crystallographic space groups in both real and reciprocal spaces. There are three types of quasicrystals: one-, two- and three-dimensional quasicrystals. In one-dimensional quasicrystals, the quasiperiodic arrangement of atoms is along one direction, while the plane perpendicular to which has a regular two-dimensional periodic arrangement. There are several sub-classes of one-dimensional quasicrystals. One typical arrangement along the unique quasiperiodic direction follows a Fibonacci sequence. In two-dimensional quasicrystals, there is a quasiperiodic two-dimensional plane with a periodic arrangement perpendicular to this plane, resulting in a true layer structure within which no translational symmetry exists. Typical two-dimensional quasicrystals include pentagonal, octagonal, decagonal, and dodecagonal quasicrystals with 5-, 8-, 10- and 12-fold symmetry respectively. In the three-dimensional quasicrystals, the atomic arrangement is quasiperiodic in all three directions, in which the icosahedral quasicrystal is the typical one. Three independent vectors used in traditional crystallography are not enough to index the diffraction peaks in quasicrystals, instead, at least four linearly independent vectors are needed [1]. The necessary n vectors span independently in n ($n > 3$) dimensional space. In other words, the quasicrystals in three-dimensional space can be constructed from a periodic crystal in a higher n dimensional space. The real structure of quasicrystal in three-dimensional physical space can be obtained by appropriate projection/section technique preserving the symmetries from n dimensional space [2]. Five miller indices are needed for describing a two-dimensional polygonal quasicrystal and six miller indices for three-dimensional icosahedral quasicrystal.

Before the discovery of alloy quasicrystals, Roger Penrose created a set of prototiles to tile a plane quasi-periodically with a strict marching rule for preserving the fivefold rotational symmetry. Later on, this famous Penrose tiling motivated much

work on the theory of quasicrystals. The third type of Penrose tiling (P3) is composed of two types of rhombi tiles with different angles. An equivalent Penrose tiling can also be constructed in three-dimensional space, resulting in icosahedral symmetry. The fivefold Penrose tiling is a geometry expression of pentagonal quasicrystals similar to other 2d symmetries including 8-, 10-, and 12-fold, as illustrated in Fig. 2.1a–c for examples [3]. The corresponding lattice vectors in reciprocal space can be obtained through projection from higher dimensional spaces as shown in the middle panel of Fig. 2.1. Four linear independent planar vectors are needed to describe the related rotational symmetry. The simulated diffraction patterns exhibit 8-, 10-, and 12-fold symmetries as shown in the bottom panel of Fig. 2.1. The fifth vector is along with the plan's normal direction.

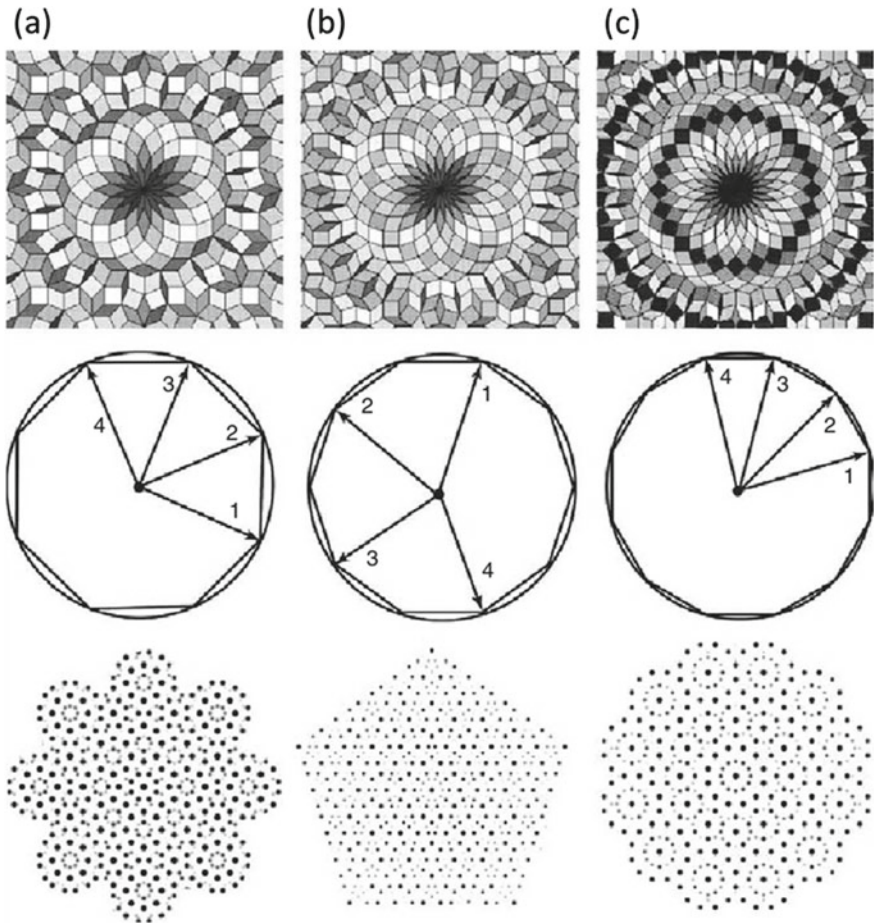


Fig. 2.1 Two-dimensional tilings (top), projection of the unit vectors of the polygonal reciprocal lattices in four-dimensional space (middle) and simulated diffraction patterns (bottom) in the quasiperiodic plane for **a** octagonal, **b** decagonal, and **c** dodecagonal quasicrystals [3]