

NANOTECHNOLOGY AND QUASICRYSTALS: FROM SELF-ASSEMBLY TO PHOTONIC APPLICATIONS

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Abstract. After providing a concise overview on quasicrystals and their discovery more than a quarter of a century ago, I consider the unexpected interplay between nanotechnology and quasiperiodic crystals. Of particular relevance are efforts to fabricate artificial functional micro- or nanostructures, as well as efforts to control the self-assembly of nanostructures, where current knowledge about the possibility of having long-range order without periodicity can provide significant advantages. I discuss examples of systems ranging from artificial metamaterials for photonic applications, through self-assembled soft matter, to surface waves and optically-induced nonlinear photonic quasicrystals.

Keywords: quasicrystals, quasiperiodic crystals, nanostructures, self-assembly, soft matter, metamaterials, nonlinear photonic crystals, surface waves, Faraday waves

1. Nanotechnology and Quasicrystals?

When organizers of the NATO Advanced Research Workshop on nanotechnology, held in St. Petersburg in June 2008, asked me to deliver a keynote lecture on quasicrystals I was certain that they had made a mistake. I have been studying quasicrystals for over 15 years and investigating nanomechanical systems for just about a decade, and although one always finds connections between different scientific fields, I had never expected such an invitation. Nevertheless, the organizers insisted and explained that they wanted to learn about the possibility of exploiting nontrivial symmetries – perhaps imitating what viruses do – and in general, learn the lesson of a scientific community that was forced by nature to keep an open mind and “think outside of the box”.

This chapter is motivated by my presentation on quasicrystals at the NATO ARW on nanotechnology in St. Petersburg. I begin in Sec. 2 by describing the discovery of quasicrystals and the scientific revolution that followed. I argue that now that the surprise has long subsided, and we are well aware of the possibility for having long-range

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order without periodicity, we are equipped with much knowledge that can be adopted in other fields – possibly nanotechnology. Particularly relevant are efforts to fabricate artificial functional micro- or nanostructures, as well as efforts to control the self-assembly of nanostructures. I give an elementary introduction to quasicrystals in Sec. 3. I then continue in Sec. 4 with an overview on the use of quasicrystalline structures in artificial metamaterials for photonic applications, and proceed in Sec. 5 to give a description of our current studies of the self-assembly of soft matter, namely micelle-forming dendrimers and polymeric stars, into quasicrystals. I make an interlude in Sec. 6 to an even softer system – parametrically-excited surface waves in fluids – that spontaneously forms quasicrystalline patterns. I finish in Sec. 8 by mentioning some dynamical properties of quasicrystals that might be more easily investigated in soft-matter quasicrystals than in solid-state quasicrystals, introducing yet another physical system, that of optically-induced nonlinear photonic quasicrystals, which we have been using to study quasicrystals.

2. The Discovery of Quasicrystals as a Modern-Day

2.1. SCIENTIFIC REVOLUTION

For almost two millennia crystallography was predominantly concerned with the external morphology of crystals. Crystallographers studied the naturally-occurring facets of crystals, which always intersect at precise and characteristic angles. It was only in the 17th century that modern crystallography was born, thanks to the brilliant idea – attributed to such great scientists as Kepler and Hooke – that crystal shapes were the result of internal *order* of “atomic” units. In his study of tilings of the plane by polygons, Kepler [1] was quick to realize that very few regular polygons – namely the triangle, the square, and the hexagon – can tile the plane without introducing overlaps or leaving holes. Yet, this observation did not deter him from constructing a well-ordered tiling of the plane, consisting of pentagons and decagons that requires some of the decagons to overlap, and leaves holes in the form of 5-fold stars – a tiling that was to be rediscovered by Penrose more than 350 years later [2, 3]. Without realizing it, Kepler had discovered some of the basic properties of aperiodic order, commenting in his own words that “*The structure is very elaborate and intricate.*”

Unfortunately, by the end of the 18th century, when Haüy began formulating the mathematical theory of crystallography [4], Kepler’s insightful drawings of aperiodic tilings with decagonal symmetry were long forgotten. Consequently, mathematical crystallography was founded upon the premise that the internal order of crystals was necessarily achieved through a periodic filling of space. Thus, crystallography treated *order* and *periodicity* synonymously, using either property interchangeably to define the notion of a *crystal*. The periodic nature of crystals was “confirmed” with the discovery of x-ray crystallography and numerous other experimental techniques throughout the 20th century. As they are more “elaborate and intricate” and less commonly found in Nature, aperiodic crystals were completely overlooked. Periodicity became the underlying paradigm, not only for crystallography itself, but also for other disciplines such as

materials science, solid state chemistry, and condensed matter physics, whose most basic experimental and theoretical tools rely on its existence.

This historical oversight was corrected with Shechtman's discovery of quasicrystals in 1982 – a discovery that sparked a *bona fide* Kuhnian scientific revolution [5], as described by Cahn [6], one of the co-authors of the announcement of the discovery, which appeared only 2 years later in 1984 [7]. In the famous Hargittai interviews [8] with all the scientists involved in the initial study of quasicrystals, Mackay is quoted as saying that

It's a discovery of a material which breaks the laws that were artificially constructed. They were not laws of nature; they were laws of the human classificatory system.

Nature had found a way of achieving order without periodicity, and Shechtman was the first to pay attention to it, and not to dismiss it as an experimental artifact, as many must have done before him. He confronted a skeptical scientific community that was unwilling to relinquish its most basic paradigm that order stems from periodicity. His biggest challenger was Pauling, one of the greatest chemists of the 20th century and a leading crystallographer of that time. In a remarkable article, suggesting an alternative description of Shechtman's icosahedral quasicrystal as multiple twinning of periodic cubic crystals – a description that not much later was shown to be incorrect [9] – Pauling [10] concluded by saying that

Crystallographers can now cease to worry that the validity of one of the accepted bases of their science has been questioned.

Today, thousands of diffraction diagrams later, compounded by high-quality experimental data – such as images from high-resolution transmission electron microscopes and atomic-resolution scanning tunneling microscopes – the existence of order without periodicity has been unequivocally established. Not only has the periodicity paradigm been questioned, as Pauling worried, it has been completely shattered. By 1992, only a decade after the discovery, the International Union of Crystallography, through its Commission on Aperiodic Crystals [11], was ready to publish a provisional definition of the term *crystal* that abolishes *periodicity*, and implies that *order* should be its replacement. The commission was not ready to give precise microscopic descriptions of all the ways in which order can be achieved. Clearly, periodicity is one way of achieving order, quasiperiodicity as in the Penrose–Kepler tiling is another, but the committee was uncertain whether there were other ways that were yet to be discovered. The Commission opted to shift the definition from a microscopic description of the crystal to a property of the data collected in a diffraction experiment. It decided on a temporary working-definition whereby a *crystal* is

any solid having an essentially discrete diffraction diagram.

Thus, crystals that are periodic are now explicitly called *periodic crystals*, all others are called *aperiodic crystals*.

The 1992 definition is consistent with the notion of long-range order – one of the basic notions of condensed-matter physics [12, 13] – dating back to ideas of Landau in which the symmetry-breaking transition from a disordered (high-symmetry) phase to an

ordered (low-symmetry) phase is quantified by the appearance of a non-zero *order parameter* – in this case, the appearance of Bragg peaks in the Fourier spectrum. Stated in plain words, long-range order – or in the context of our current discussion, long-range positional order – is a measure of the correlations between the positions of atoms in distant regions of the material.

The 1992 definition was left sufficiently vague so as not to impose unnecessary constraints on any further study of crystallinity. Indeed, the discovery inspired a renaissance in crystallography, and has made a great impact on mathematics [14], as can be seen by an ever increasing number of books [15–18]. Much effort has been invested in studying the characteristics of order, as well as in the development of diffraction theory [19–21], once it was realized that periodicity was not a necessary condition for order and for the appearance of Bragg peaks in a diffraction spectrum. In particular, Baake and several co-workers [22–26] have performed a systematic study whose purpose is to characterize which distributions of matter diffract to produce a pure point component in their spectrum, and thus can qualify as possessing long-range order. Sufficient progress has already been made, that we are now ready to complete the paradigm shift and adopt a permanent definition of crystal that is firmly based on the notion of *order* [27].

In October 2007 the quasicrystal community – consisting of mathematicians, physicists, chemists, materials scientists, surface scientists, and even photonics engineers – celebrated the 25th anniversary of the discovery in a “Silver Jubilee” conference [28]. Today, the science of quasicrystals, with its growing number of textbooks [15–18, 29–33], is a mature science. Old paradigms have been carefully transformed into new ones [34]; definitions have been changed [35]; space-group theory has been generalized to quasicrystals using two alternative approaches [36–39], and even extended to treat novel long-range order possessing color [40] or magnetic symmetry [41, 42]); and many fundamental problems – including Bak’s famous question: “Where are the atoms?” [43] – are finding their solutions [44, 45]. Nevertheless, other important questions have remained unanswered to this day. Many of these – such as the electronic and other physical properties of quasicrystals [46], the surface science of quasicrystals [47], and the importance of the phason degree of freedom [48] – were hotly debated at the “Silver Jubilee” conference [28], and continue to drive us forward. One particularly interesting set of questions, and the focus of this chapter, deals with metamaterials and soft-matter quasicrystals – the newly added members of the quasicrystal family.

We know today that quasicrystals are more common than one had originally expected. Scores, or even hundreds, of binary and ternary *metallic alloys* are known to form quasicrystalline phases [49] – mostly with icosahedral or decagonal point-group symmetry – and more are continuously being discovered. Nevertheless, it is only in the last few years that quasicrystals have been discovered (independently) in two different *soft-matter* systems: micelle-forming dendrimers [50–52] and three-armed star block copolymers [53–55]. These newly discovered *soft quasicrystals* not only provide exciting alternative experimental platforms for the basic study of quasiperiodic long-range order, but also hold the promise for new applications based on self-assembled

nanomaterials [56–58], with unique electronic or photonic properties that take advantage of the quasiperiodicity, which is relevant to our focus here.

The current emphasis in the study of soft-matter quasicrystals is to find an explanation for their thermodynamic stability, and thus learn how to control their self-assembly. To this date, soft quasicrystals have been observed only with dodecagonal point-group symmetry. Their source of stability is therefore likely to be different from their solid-state siblings, yet a good understanding of the stability of one quasiperiodic system may help to understand the stability of the other. In what follows I shall review our initial understanding of what might be the source of stability of soft quasicrystals, while providing a concise background on the subject. I will try to emphasize the important relations between the variety of different physical and chemical systems that form quasicrystalline phases – atomic quasicrystals, soft quasicrystals, surface waves, and also artificially-produced structures and metamaterials.

3. Quasicrystals – Terminology and General Framework

Let us consider a scalar function $\rho(\mathbf{r})$ that describes the electronic density or the ionic potential of a material. The Fourier transform of a quasiperiodic density $\rho(\mathbf{r})$ has the form

$$\rho(\mathbf{r}) = \sum_{\mathbf{k} \in L} \rho(\mathbf{k}) e^{i\mathbf{k} \cdot \mathbf{r}}, \quad (1)$$

where the (reciprocal) lattice L is a finitely generated \mathbb{Z} -module, i.e. it can be expressed as the set of all integral linear combinations of a finite number D of d -dimensional wave vectors, $\mathbf{b}^{(1)}, \dots, \mathbf{b}^{(D)}$. In the special case where D , called the rank of the crystal, is equal to the physical dimension d , the crystal is periodic. We refer to all quasiperiodic crystals that are not periodic as “quasicrystals”.¹ This term was first coined by Levine and Steinhardt [59, 60] in the first of a series of important theoretical papers that were published in the 1980s.

It is useful to introduce a physical setting based on the notion of symmetry breaking that was mentioned earlier [12, 13, 62]. Let us assume that the quasiperiodically-ordered state, described by $\rho(\mathbf{r})$, is a symmetry-broken stable ground state of some generic free energy F , invariant under all translations and rotations in \mathbb{R}^d . This is the same as saying that the physical interactions giving rise to the quasicrystal are themselves translationally and rotationally invariant, and that the ground state breaks this symmetry. The free energy F is a functional of $\rho(\mathbf{r})$, which in Fourier space takes the general form

¹ Some older texts require crystals to possess so-called “forbidden symmetries” in order to be regarded as quasicrystals. It is now understood that such a requirement is inappropriate. See Ref. [35] for details, and Ref. [61] for examples of square and cubic quasicrystals.

$$F\{\rho\} = \sum_{n=2}^{\infty} \sum_{\mathbf{k}_1, \dots, \mathbf{k}_n} A(\mathbf{k}_1, \dots, \mathbf{k}_n) \rho(\mathbf{k}_1) \cdots \rho(\mathbf{k}_n). \quad (2)$$

Based on the idea of such a generic free energy, Rokhsar, Wright, and Mermin [36] introduced the notion of *indistinguishability*, namely that two functions $\rho(\mathbf{r})$ and $\rho'(\mathbf{r})$ are indistinguishable if a generic free energy cannot distinguish between them and assigns them both the same value. It then follows [36–38] that $\rho(\mathbf{r})$ and $\rho'(\mathbf{r})$ are indistinguishable if and only if

$$\forall \mathbf{k} \in L: \quad \rho'(\mathbf{k}) = e^{2\pi i \chi(\mathbf{k})} \rho(\mathbf{k}), \quad (3)$$

where $\chi(\mathbf{k})$, called a *gauge function*, has the property that $\chi(\mathbf{k}_1 + \mathbf{k}_2) \equiv \chi(\mathbf{k}_1) + \chi(\mathbf{k}_2)$ whenever \mathbf{k}_1 and \mathbf{k}_2 are in L , where ‘ \equiv ’ denotes equality to within an additive integer.

Gauge functions are useful in describing the relations between the different symmetry-broken ground states of F . Dräger and Mermin [63] showed that gauge functions form a vector space V^* of all real-valued linear functions on the lattice L , and because L has rank D , V^* is a D -dimensional vector space over the real numbers. The space V^* contains, as a subspace, all the integral-valued linear functions on L . This subset, which has the algebraic structure of a rank- D \mathbb{Z} -module (just like L itself) is denoted by L^* . Gauge functions in L^* leave the ground-state density invariant. Gauge functions that belong to the quotient space V^*/L^* take the ground state described by ρ into a different, yet indistinguishable, ground state described by some other density function ρ' . Thus, one can parameterize all the related symmetry-broken ground states of F on a simple D -torus – the order parameter space c .

Different, yet indistinguishable, ground states may also be related by rotations $g \in O(d)$. In this case ρ' in (3) is simply a rotated version of ρ , and for each such rotation g there is a special gauge function φ_g , called a *phase function*, satisfying

$$\forall \mathbf{k} \in L: \quad \rho(g\mathbf{k}) = e^{2\pi i \varphi_g(\mathbf{k})} \rho(\mathbf{k}). \quad (4)$$

The set of all rotations satisfying (4) forms the point group of the crystal, and along with the corresponding phase functions completely characterizes its space group [36–38, 40–42]. Unlike periodic crystals, quasicrystals are not restricted in the order of their rotational symmetry. The point-group condition (4) is applicable to operations of any order (as long as the rank of the crystal is finite). Thus, in general, g may be an n -fold rotation of any order n .

4. Exploiting Quasicrystalline Order in Artificially Constructed Metamaterials

Interesting applications are starting to emerge lately that take advantage of quasiperiodic long-range order in metamaterials, or artificially constructed quasicrystals [64]. Two

main features distinguish quasicrystals from periodic crystals in the practical sense of using them as metamaterials. The first and more obvious is the relaxation of any symmetry constraints. In dealing with quasicrystals for over a quarter of a century we have learned how to design structures with axes of symmetry of arbitrarily high order. Of course, as the order of symmetry increases, so does the rank of the crystal and therefore its complexity. Nevertheless, simple rank-4 2-dimensional structures already allow one to construct structures with axes of 5-fold, 8-fold, 10-fold, and 12-fold symmetry – a substantial improvement over the limited 2-fold, 3-fold, 4-fold, and 6-fold axes possible with periodic crystals. Most applications of quasicrystalline metamaterials to date are thus based on this notion. These are mostly *linear photonic crystals*, where quasiperiodic modulations of the index of refraction of different materials are used in order to engineer their optical response. These applications take advantage of the fact that there are no restrictions on the order of the rotational symmetry in order to obtain nearly-isotropic photonic band gaps [65, 66]. Dodecagonal (12-fold) quasicrystals are particularly useful as they are at the same time quite simple (the rank is only 4) yet the dodecagon is a far better approximation of a circle than the hexagon, which is the best one can achieve with periodic photonic crystals. Initial work is also carried out with *phononic quasicrystals* for controlling the propagation of sound waves [64].

The second feature of quasicrystals, useful for metamaterial applications, is the complete relaxation of any constraints on the positions of Bragg peaks in their diffraction diagrams. One may design quasiperiodic metamaterials in which the Bragg peaks are placed at predetermined positions in Fourier space. We have exploited this idea in the *nonlinear* optical domain [67–69], where recent technological progress has enabled to modulate the second-order nonlinear susceptibility with micron-scale resolution in various materials, such as ferroelectrics, semiconductors, and polymers. In these *nonlinear photonic crystals* the modulation can be achieved by planar techniques, thereby offering either one or two dimensions for modulation. Moreover, there are no photonic band gaps in these metamaterials, because the first-order susceptibility, and hence the refractive index, remain constant. We emphasize that the advantage of using quasicrystals in this case is not in their arbitrarily-high symmetry, but rather in the fact that there is no restriction on the combinations of wave vectors that may appear in their reciprocal lattices (provided that the symmetry of the quasicrystal is not of particular importance [38, 70]).

The novel optical devices that we have been developing are based on materials that facilitate the nonlinear interaction between light waves in the form of three-wave mixing. These are processes in which two incoming waves of frequencies ω_1 and ω_2 interact through the quadratic dielectric tensor $\chi^{(2)}$ of the material to produce a third wave of frequency $\omega_3 = \omega_1 \pm \omega_2$; or the opposite processes in which a single wave spontaneously breaks up into two. Three-wave mixing is severely constrained in dispersive materials, where $\omega(\mathbf{k})$ is not a linear function, because the interacting photons must also conserve their total momentum. Even the slightest wave-vector mismatch $\Delta\mathbf{k} = \mathbf{k}_1 \pm \mathbf{k}_2 - \mathbf{k}_3$ appears as an oscillating phase that averages out the outgoing wave, giving rise to the so-called “phase-matching problem.” We have explained how one could fully solve the most general phase-matching problem using

well-known ideas from the theory of quasicrystals [67]. The solution is based on the idea that in crystals, whether periodic or not, continuous translation symmetry is broken, as described above in Sec. 3. As a consequence, momentum conservation is replaced by the less-restrictive conservation law of crystal momentum whereby momentum need only be conserved to within a wave vector from the reciprocal lattice of the crystal. The fabrication of an efficient frequency-conversion device is therefore a matter of *reciprocal-lattice engineering* – designing an artificial crystal, from the quadratic dielectric field of the material $\chi^{(2)}(\mathbf{r})$, whose reciprocal lattice contains any desired set of mismatch wave vectors $\Delta\mathbf{k}^{(j)}$, $j = 1 \dots N$, required for phase matching any arbitrary combination of N three-wave mixing processes.

The idea of using a 1-dimensional periodic modulation of the relevant component of the quadratic dielectric tensor, for the purpose of phase matching a single three-wave process, was suggested already in the early 1960s [71–73], and is termed “quasi-phase matching”. Since then this approach has been generalized using more elaborate 1-dimensional [74–76] and 2-dimensional [77–80] designs, but only as *ad hoc* solutions for multiple processes. We have demonstrated that engineering the reciprocal lattice of a nonlinear photonic quasicrystal to contain any desired set of mismatch vectors – a task that 25 years of research in quasicrystals have taught us how to solve – provides the most general solution for the long-standing problem of multiple phase-matching [67]. We described elsewhere [68, 69] a number of novel optical devices that have actually been fabricated using these ideas, and tested experimentally. These devices attest to the general nature of the quasicrystal-based solution to the multiple phase-matching problem.

5. Towards Self-Assembly of Quasicrystalline Nanostructures – The Recent Discovery of Soft-Matter Quasicrystals

An important development accrued recently with the experimental discovery that even soft matter can self-assemble into structures with quasiperiodic long-range order.² In one case, dendrimers that assume a conical shape assemble into micelles, which then pack to form a perfect dodecagonal (12-fold) quasicrystal [50–52]. In another case, *ABC* star-shaped block terpolymers – in which the length ratios of the three arms, B/A and C/A , can be chemically-controlled – assemble into a host of 2-dimensional columnar structures, one of which is, again, a dodecagonal quasicrystal [53–55]. This phase has also been reproduced numerically using lattice Monte Carlo simulations [87]. A similar square-triangle tiling has also been observed in a liquid crystal composed of T-shaped molecules [88], which forms yet a third soft system which may potentially self-assemble into a dodecagonal quasicrystal. The characteristic length of the basic building blocks

² For the sake of historical accuracy, it should be noted that at some point the blue phase III of liquid crystals, also known as the “blue fog”, was thought to have icosahedral quasicrystalline order [81, 82], but this eventually turned out not to be the case [83, 84]. Also, incommensurate helical twist-grain-boundary phases are known to exist in smectic liquid crystals [85, 86], but the quasiperiodic order in this case is essentially only along the 1-dimensional screw axis.

ranges in these systems from about 10 to about 100 nm – 2 to 3 orders of magnitude greater than the atomic length scales found in solid-state quasicrystals. This property of soft quasicrystals is what will potentially make them useful as functional self-assembled nanomaterials [56–58], and at the same time as a new experimental platform for detailed – real-space and real-time – study of quasiperiodic long-range order.

Investigations of these new soft members of the quasicrystal family of materials, are only at their infancy. For example, even the space groups of the observed phases have not been determined, although from the diffraction patterns of the dendrimer liquid crystals given by Zeng et al. [50, 51] it seems that they have a 12-fold screw axis, and therefore, most likely, the nonsymmorphic space group $P12_6/mcm$ [37]. More generally, the same questions [46–48] concerning the thermodynamic stability, the role of clusters in formation and dynamics, and the importance of phasons, apply to soft quasicrystals as they do to hard quasicrystals. Yet the answers may be more tractable (albeit possibly different as the systems are quite different). Thus, the study of soft quasicrystals will clearly have implications well beyond the limits of the specific soft systems that have been discovered so far, and is likely to promote the fundamental understanding of quasicrystals in general. Fortunately, the study of soft quasicrystals is happening at a point in time when the science of quasicrystals is ready and mature to tackle these newly discovered systems. We are no longer taken by surprise whenever a new chemical or physical system exhibits quasicrystalline structure. We are prepared with the appropriate tools to study and explore it, and hopefully also to exploit it for the control of the self-assembly of useful nanomaterials.

6. Insights from an Even Softer System – Quasicrystalline Surface Waves

Motivated by experiments with parametrically-excited surface waves (Faraday waves), exhibiting dodecagonal quasiperiodic order [89], Lifshitz and Petrich [90] developed some years ago a model for describing the pattern-forming dynamics of a 2-dimensional field in which two length scales undergo a simultaneous instability. This model is an extension of the Swift–Hohenberg equation [91], which is used for describing a variety of different pattern-forming systems [92]. Its dynamics is relaxational, $\partial_t \rho = -\delta F / \delta \rho$, driving a 2-dimensional field $\rho(x, y, t)$ towards the minimum of an “effective free energy” (2),

$$F_{LP}\{\rho\} = \int dx dy \left\{ -\frac{1}{2} \varepsilon \rho^2 + \frac{1}{2} [(\nabla^2 + 1)(\nabla^2 + q^2)\rho]^2 - \frac{1}{3} \alpha \rho^3 + \frac{1}{4} \rho^4 \right\}, \quad (5)$$

yielding a dynamical equation of the form

$$\partial_t \rho = \varepsilon \rho - (\nabla^2 + 1)^2 (\nabla^2 + q^2)^2 \rho + \alpha \rho^2 - \rho^3 \quad (6)$$

It essentially mimics the dynamics of a generic 2-dimensional material in search of its ground state, and therefore offers us important insight and a good starting point for

our current investigation of soft quasicrystals. A Java simulation of the dynamical equation (6), starting from random initial conditions, and arriving at a quasicrystalline pattern can be found at http://www.its.caltech.edu/~mcc/Patterns/Demo4_6.html.

The Lifshitz–Petrich free energy F_{LP} is indeed generic, imposing only two requirements on a material, described by a 2-dimensional density $\rho(x, y, t)$: (a) The existence of two characteristic length scales, whose ratio is given by the parameter q ; and (b) The existence of effective 3-body interactions, whose importance is given by the relative strength of the parameter α . In [90] we were able to show analytically (using standard methods [93, 94]), and demonstrate numerically, that if q is chosen around $2 \cos(\pi/12) = \sqrt{2 + \sqrt{3}} \cong 1.932$ one can obtain a ground state with quasiperiodic long-range order and dodecagonal symmetry, yet no choice of q yields globally-stable ground states with octagonal or decagonal symmetry. The latter two have insufficient triplets of wave vectors in the Fourier Lattice L [Eq. (1)] that add up to zero to overcome the cost of additional density modes, as compared with the hexagonal state. Thus, in 2-dimensions, the requirements of two length scales and 3-body interactions are sufficient to stabilize dodecagonal quasicrystals, but insufficient to stabilize octagonal or decagonal quasicrystals. This raises the possibility that the fact that the soft quasicrystals discovered to date are all dodecagonal, may be accounted for using a free energy similar to F_{LP} . Note that for hard quasicrystals the situation is different – decagonal quasicrystals are thermodynamically stable whereas octagonal and dodecagonal quasicrystals are believed to be metastable – indicating that the stabilization mechanism for soft quasicrystals might be quite different from that of hard quasicrystals.

7. Validity of Density-Wave Theories of Quasicrystals

At the outset, as we argue in more detail elsewhere [95], the experimental soft systems in which quasicrystalline order has been observed seem to satisfy the basic assumptions of the Lifshitz–Petrich theory described in Sec. 6. The asymmetric and heterogeneous structure of the star polymers and dendrimers will most likely require more than one length scale for an appropriate coarse-grained description.³ Their ultra-soft repulsion and resulting strong inter-penetration [99–105] imply that 3-body interactions should be significant [106]. Thus, we expect that studies that we are currently undertaking will yield functionals similar in nature to F_{LP} of Eq. (5). Significant differences may emerge, nonetheless, as the systems considered here are 3-dimensional and differ in their microscopic structure. For instance, two order parameters rather than one might be required [55], which could potentially allow point-group symmetries other than dodecagonal to be observed.⁴

Another key insight can be drawn from a recent theoretical observation, according to which dispersions of soft, fuzzy, particles are essentially different in their thermodynamics

³ Indeed, coarse-grained free energies previously used for amphiphilic self-assembly [96] involve more than one characteristic length scale due to the asymmetry of the molecules and the resulting tendency to form curved interfaces.

⁴ Models with two order parameters were suggested also for hard quasicrystals [97] and pattern-forming systems [98], yielding additional quasicrystalline ground-state symmetries.

from those of hard particles [107, 108]. The overlap of the soft “coronas” surrounding the particles leads to a driving force acting to minimize their interfacial area, in analogy with foams. Consequently, unusual liquid-crystalline structures can be stabilized in systems of soft spheres [107–110]. Both star polymers and flexible dendrimers fall into this fuzzy category [111, 112], yet they may be highly aspherical. Likos et al. [104] have also shown that stars and flexible dendrimers have the same kind of soft pair potentials. We thus expect such considerations of interfacial-area minimization to become highly relevant in the study of soft quasicrystals.

A 3-dimensional version of an LP-like free energy may remind the reader of the early attempts by Kalugin et al. [113, KKL], who extended the model of Alexander and McTague [114], using density-wave theories to establish that the icosahedral quasicrystal has lower free energy than the competing *bcc* phase. Narasimhan and Ho [115, NH] managed to show in their model that there are regions in parameter space in which a dodecagonal quasicrystal is favored and other regions in which a decagonal quasicrystal is favored. These attempts were eventually discontinued after it was shown by Gronlund and Mermin [94] that the addition of a quartic term to the cubic free energy of KKL reverses the outcome of the calculation, establishing the *bcc* phase as the favored one. For hard crystals it is unclear where to truncate the density-wave expansion of the free energy and whether such a truncation is fully justified. As we discussed in [95], for the soft systems considered here the truncation of the expansion should be more valid. We are therefore in a position now to reexamine some of the old conclusions, based on density-wave theories of quasicrystals, as they are likely to apply to *soft* quasicrystals. Roan and Shakhnovich [116] performed such a stability study for the case of icosahedral order in diblock copolymers and concluded that such order is only metastable. Nevertheless, we are encouraged by the old results of NH who established the stability of dodecagonal, as well as decagonal, quasicrystals within the same model.

8. Dislocation and Phason Dynamics – From Soft to Photonic Quasicrystals

Valuable knowledge about the nature of quasiperiodic order, important also for the control of its self-assembly, can be obtained by studying its topological defects [12, 13, 117], and its low-energy collective excitations – in particular those associated with the phason degrees of freedom. Much like phonons, phasons are low-energy excitations of the quasicrystal, only that instead of slightly shifting the atoms away from their equilibrium positions, the relative positions of atoms are interchanged. Their existence stems directly from the fact that the dimension D of the order parameter space V^*/L^* is greater than the physical dimension d . Thus, in addition to d independent (acoustic) phonon modes there are $D - d$ independent phason modes.

The existence of phasons as fundamental degrees of freedom, affecting the physical behavior of quasicrystals, has been clearly established over the years. Their role in a dynamical density-wave theory of quasicrystals was developed in a series of papers [9, 118–124] immediately following the announcement of the discovery of quasicrystals. Phasons have been observed in numerous experiments, whether directly or indirectly,

throughout the past two decades [125–129], and are still a source of interesting analytical puzzles [130] and ongoing debate [48].

We have recently begun investigating the motion of dislocations and the dynamics of phasons in the dodecagonal ground state of the LP equation [131]. We are studying, both analytically and numerically, such questions as the climb velocity of dislocations under strain, the pinning of dislocations by the underlying quasiperiodic structure under conditions of weak diffusion, and the relaxation of phason strain as two dislocations of opposite topological sign merge and annihilate each other. These studies are impossible to conduct with either Faraday waves or hard atomic quasicrystals. Thus soft quasicrystals, with their 10 to 100 nm length scales, may become one of the first natural experimental systems to provide real quantitative microscopic answers regarding the dynamics of the fundamental degrees of freedom in a quasicrystal – defects and low-energy excitations.

In the meantime we have embarked on the study of these unique dynamical degrees of freedom in a fascinating new artificial form of quasicrystalline medium – an optically-induced nonlinear photonic quasicrystal – which we have recently demonstrated [132]. In this system beams of light interact nonlinearly by changing the index of refraction of a photorefractive material. Their dynamics is governed by a different type of equation – the so-called nonlinear Schrödinger equation. Nevertheless, it is capable of stabilizing structures with quasicrystalline order where the typical distance between crystal sites is 15 to 30 μm . This has allowed us to study the microscopic dynamics of dislocations [132] as well as phasons [133]. These artificial systems already provide useful information regarding the dynamics of fundamental degrees of freedom in quasicrystals. Similar investigations of soft quasicrystals, should provide valuable insight into their physical nature, as well as that of all quasicrystals, regardless of the physical or chemical system in which they are realized. This insight will be most valuable in trying to design and control the self assembly of quasiperiodic nanomaterials.

Acknowledgments: I would like to thank Alexey Madison, Yuri Magarshak, and Sergey Kozirev for inviting me to St. Petersburg, where I was encouraged to stress the connections between my different fields of research – nanotechnology and quasicrystals. I would like to thank Haim Diamant and Kobi Kan with whom I am currently trying to understand the stability of soft quasicrystals and how one could control their self assembly. I thank Gilad Barak with whom I have studied the dynamics of dislocations and phasons in the Lifshitz–Petrich equation. I thank Ady Arie, Alon Bahadad, and Noa Voloch with whom I have been developing the use of nonlinear photonic quasicrystals for the purpose of frequency conversion. Finally, I would like to thank Barak Freedman and Mordechai Segev with whom I have been studying the dynamics of optically-induced nonlinear photonic quasicrystals. This work is funded by the Israel Science Foundation under Grant No. 684/06.

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