

Soft quasicrystals – Why are they stable?

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In the last two years we have witnessed the exciting experimental discovery of soft matter with nontrivial quasiperiodic long-range order – a new form of matter termed a *soft quasicrystal*. Two groups have independently discovered such order in soft matter: Zeng *et al.* in a system of dendrimer liquid crystals; and Takano *et al.* in a system of ABC star-shaped polymers. These newly discovered soft quasicrystals not only provide exciting platforms for the fundamental study of both quasicrystals and of soft matter, but also hold the promise for new applications based on self-assembled nanomaterials with unique physical properties that take advantage of the quasiperiodicity, such as complete and isotropic photonic band-gap materials. Here we provide a concise review of the emerging field of soft quasicrystals, suggesting that the existence of two natural length-scales, along with three-body interactions, may constitute the underlying source of their stability.

1. Background

The discovery of quasicrystals by Shechtman more than two decades ago [1] signalled the beginning of a remarkable scientific revolution [2], in which some of the most basic notions of condensed matter physics have undergone a thorough reexamination. Today, the science of quasicrystals, with its growing number of textbooks, is in its adolescence. Old paradigms are being carefully transformed into new ones [11]; definitions are being changed [12]; space-group theory has been generalized to quasicrystals using two alternative approaches [13–16], and even extended to treat novel long-range order possessing colour [17] or magnetic symmetry [18, 19]; and many fundamental problems – including Bak’s famous question: ‘Where are the atoms?’ [20] – are gradually finding their solutions. Nevertheless, other questions that are equally important have remained unanswered to this day. Some of these – such as the stabilization of quasicrystals, the role of clusters, and the importance

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§Early papers in the field were collected by Steinhardt and Ostlund [3]; for an introductory textbook on quasicrystals, see [4]; for collections of reviews on the physical properties of quasicrystals, see [5, 6]; for an introductory textbook on the mathematics of quasicrystals, see [7]; for detailed reviews on the mathematics of quasicrystals, see [8, 9]; for a detailed overview on the applications of quasicrystals, see [10].

of the phason degree of freedom – were hotly debated at a recent International Conference on Quasicrystals [21].

We know today that quasicrystals are more common than one had originally expected. Scores of binary and ternary *metallic alloys* are known to form quasicrystalline phases [22] – mostly with icosahedral or decagonal point-group symmetry – and more are being discovered all the time. Nevertheless, it is only in the last couple of years that quasicrystals have been discovered (independently) in two different *soft-matter* systems: dendrimers [23–25] and possibly star block copolymers [26–28]. 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 [29–31], with unique electronic or photonic properties that take advantage of the quasiperiodicity. One example is the isotropic photonic band gaps that have been demonstrated in artificially-constructed octagonal [32] and dodecagonal [33] quasicrystals.

The first step in a theoretical study of soft quasicrystals would be to provide an explanation for their thermodynamic stability. 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. The purpose of this article is to propose initial suggestions as to the source of stability of soft quasicrystals, while providing a concise background on the subject.

2. 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 $\rho(\mathbf{r})$ (we shall assume here that the transform always exists) 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’[†].

It is useful to introduce a physical setting based on the notion of symmetry breaking [35–37]. 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 \mathcal{F} , invariant under all translations and rotations in \mathbb{R}^d . This is the same as saying that

[†]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. [12] for details and Ref. [34] for examples of square and cubic quasicrystals.

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 \mathcal{F} is a functional of $\rho(\mathbf{r})$, which in Fourier space takes the general form

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

Based on the idea of such a generic free energy, Rokhsar, Wright, and Mermin [13] 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 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 \mathcal{F} . Dräger and Mermin [38] 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 \mathcal{F} on a simple D -torus – the order parameter space V^*/L^* .

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

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

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 [13–15, 17–19].

3. Ordered soft matter

A wide variety of materials in physical-chemical and biological systems lie in the intermediate region between simple liquids and crystalline solids. Examples include liquid crystals, suspensions, emulsions, polymer solutions or melts, surfactant solutions, and biomaterials [39]. These materials, termed *complex fluids*, *structured fluids*, or *soft condensed matter*, are mostly viscoelastic liquids, which contain a certain degree of inner structure. Unlike simple fluids or solids, where either

entropy (in the former) or interactions (in the latter) dominate, the behaviour of soft matter is determined by an intricate interplay between interactions and thermal fluctuations. Consequently, soft-matter systems exhibit remarkably rich structures and dynamics, which have been the subject of extensive experimental and theoretical research during the past several decades [39–43].

Soft materials may possess long-range or quasi-long-range order [44, 45]. As was first demonstrated by Onsager [46], such liquid-crystalline phases can be obtained in systems as simple as molecular liquids whose molecules have anisotropic shapes. In more complex, self-assembling systems, liquid-crystalline phases are usually made of intermediate structures, which form as a result of incompatible molecular groups linked together by chemical bonds. For example, amphiphilic molecules (surfactants) are composed of hydrophilic and hydrophobic groups, leading, in aqueous solution, to the formation of mesoscopic structures such as micelles or fluid membranes [40–42]. These structures, at sufficiently high concentration, organize into various ordered phases [42, 43]. Another important example is block copolymers – macromolecules composed of two or more chemically bonded, chain-like blocks [47–51]. The incompatibility of the different blocks gives rise to a variety of ordered structures in melts and in solutions. The main advantage of block copolymers as self-assembling systems is the relative ease at which the sizes and properties of the different blocks (and, thus, the resulting mesoscopic structures) can be controlled. Block copolymers, like homopolymers, can be linear or branched. Two branched architectures which can be well controlled, and thus have been drawing considerable attention recently, are star polymers [52–63], where the chains are joined in one point, and dendrimers [60, 64–67], having a tree-like structure.

Phenomenological models based on generic free energies similar to (2) have been very successful in describing phase diagrams and transitions in soft matter [43–45, 50, 51, 68, 69]. In such models one identifies an order parameter, which may be a scalar function – as in equation (2) – or a vector function, and formulates, based on symmetry, a free-energy functional as an expansion in the order parameter and its gradients. Such an expansion is generally expected to become a good approximation close to a continuous phase transition, where variations in the order parameter occur over large length scales. The reason for the success of this approach for soft systems, in particular, lies in their intermediate, mesoscopic building blocks, which are significantly larger than the atomic scale. For example, spatial variations of the order-parameter in block copolymer phases must occur over lengths larger than a polymer block, which may be many nanometers. As a result, composition variations and interfaces are smoother in soft materials than in crystalline solids, rendering a long-wavelength gradient expansion a much better approximation. The situation becomes even better in the case of flexible star polymers and dendrimers – which are of particular relevance here – as the effective pair potentials in these systems have an ‘ultra-soft’ short-range repulsion [54–57, 60, 62, 63, 67].

4. Recent discovery of soft-matter quasicrystals

Our current discussion is motivated by the recent experimental discovery that soft matter can self-assemble into structures with quasiperiodic

long-range order[†]. In one case, dendrimers that assume a conical shape assemble into micelles, which then pack to form a perfect dodecagonal (12-fold) quasicrystal [23–25]. 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 two-dimensional columnar structures. One of these, composed of square and triangle motifs (tiles), is suspected as being a dodecagonal quasicrystal, or at least a periodic approximant of such a quasicrystal, which under minor parameter tweaking may indeed form a perfect quasicrystal [26–28]. This phase has also been reproduced numerically using lattice Monte Carlo simulations by Dotera and Gemma [76]. A similar square-triangle tiling has also been observed in a liquid crystal composed of T-shaped molecules [77], which forms yet a third soft system which may potentially self-assemble into a dodecagonal quasicrystal. The characteristic length of the basic building blocks ranges in these systems from ~ 10 to ~ 100 nanometers – 2 to 3 orders of magnitude greater than the atomic length scales found in hard quasicrystals. This property of soft quasicrystals is what will potentially make them useful as functional self-assembled nanomaterials, and at the same time as a new experimental platform for detailed – real-space and real-time – study of quasiperiodic long-range order.

Very little is known at this point about these soft quasicrystals. 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.* [23, 24] it seems that they have a 12-fold screw axis, and therefore, most likely, the nonsymmorphic space group $P12_6/mcm$ [14]. More generally, the same questions [21] concerning the mechanism of stabilization, 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.

5. Why are soft quasicrystals stable? – insight from the Lifshitz–Petrich equation

Motivated by experiments with parametrically-excited surface waves (Faraday waves), exhibiting dodecagonal quasiperiodic order [78], Lifshitz and Petrich [79] developed a model for describing the pattern-forming dynamics of a two-dimensional field in which two length scales undergo a simultaneous instability. This model is an extension of the Swift–Hohenberg equation [80], which is used for describing a variety of different pattern-forming systems [81]. Its dynamics

[†]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 [70, 71], but this eventually turned out not to be the case [72, 73]. Also, incommensurate helical twist-grain-boundary phases are known to exist in smectic liquid crystals [74, 75], but the quasiperiodic order in this case is essentially only along the one-dimensional screw axis.

are relaxational, $\partial_t \rho = -\delta \mathcal{F} / \delta \rho$, driving a two-dimensional field $\rho(x, y, t)$ towards the minimum of an ‘effective free energy’ (2),

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

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 two-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.

The Lifshitz–Petrich free energy \mathcal{F}_{LP} is indeed generic, imposing only two requirements on a material, described by a two-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 three-body interactions, whose importance is given by the relative strength of the parameter α . In [79] we were able to show analytically (using standard methods [44, section 4.7] and [82]), and demonstrate numerically, that if q is chosen around $2 \cos(\pi/12) = \sqrt{2 + \sqrt{3}} \simeq 1.932$ 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 (equation (1)) that add up to zero to overcome the cost of additional density modes, as compared with the hexagonal state. Thus, in two dimensions, the requirements of two length scales and three-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 \mathcal{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.

6. Future directions

6.1. Free-energy functionals and stability analysis

At the outset, the experimental soft systems in which quasicrystalline order has been observed seem to satisfy the basic assumptions of the Lifshitz–Petrich theory described in section 5. 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[†]. Their ultra-soft repulsion and resulting

[†]Indeed, coarse-grained free energies previously used for amphiphilic self-assembly [43] involve more than one characteristic length scale due to the asymmetry of the molecules and the resulting tendency to form curved interfaces.

strong inter-penetration [54–57, 60, 62, 63, 67] imply that three-body interactions should be significant [59]. Thus, we expect that studies that we are currently undertaking will yield functionals similar in nature to \mathcal{F}_{LP} of equation (5). Significant differences may emerge, nonetheless, as the systems considered here are three-dimensional and differ in their microscopic structure. For instance, two order parameters rather than one might be required [28], which could potentially allow point-group symmetries other than dodecagonal to be observed[†].

A three-dimensional version of an LP-like free energy may remind the reader of the early attempts by Kalugin, Kitaev, and Levitov [85, KKL], who extended the model of Alexander and McTague [86], to establish that the icosahedral quasicrystal has lower free energy than the competing *bcc* phase. Narasimhan and Ho [87, NH] managed to show in their model that there are regions in parameter space in which a dodecagonal quasicrystal is favoured and other regions in which a decagonal quasicrystal is favoured. These attempts were eventually discontinued after it was shown by Gronlund and Mermin [82] 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 favoured one. For hard crystals it is unclear where to truncate the expansion of the free energy and whether such a truncation is fully justified. As has been mentioned in section 3, for the soft systems considered here the truncation of the expansion should be more valid. We therefore intend to reexamine some of the old conclusions when considering the stability of *soft* quasicrystals based on generic free energy considerations. Roan and Shakhnovich [88] performed such a 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.

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 from those of hard particles [89, 90]. 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 [89–92]. Both star polymers and flexible dendrimers fall into this fuzzy category [93, 94], yet they may be highly aspherical. Likos *et al.* [67] 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 upcoming study of soft quasicrystals.

6.2. Dislocation and phason dynamics

Valuable knowledge about the nature of quasiperiodic order can be obtained by studying its topological defects [35, 37, 95], and its low-energy collective excitations – in particular those associated with the phason degrees of freedom. Much like

[†]Models with two order parameters were suggested also for hard quasicrystals [83] and pattern-forming systems [84], yielding additional ground-state symmetries.

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 changed. 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 exact role of phasons in the stabilization and the dynamics of quasicrystals is not fully-understood [21], yet their presence is unequivocally-detected in numerous types of experiments. Edagawa *et al.* [96, 97] have recently managed to observe the real-time thermal fluctuations of single phason-flips in a solid-state quasicrystal – a remarkable achievement that requires a very sophisticated experiment. Soft quasicrystals, with their larger length scales, may be much better suited for real-time investigation of phason dynamics.

We have recently begun investigating the motion of dislocations and the dynamics of phasons in the dodecagonal ground state of the LP equation [98, 99]. 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. We have also used similar theoretical procedures in the analysis of defect dynamics in a real experimental nonlinear photonic quasicrystal [100]. Similar investigations of more realistic models of soft quasicrystals, or on the soft systems themselves, should provide valuable insight into their physical nature.

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