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Discussion of phasons in quasicrystals and their dynamics

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Phasons are a type of excitation peculiar to quasicrystals and other incommensurate phases arising as a consequence of their spatial quasiperiodicity. Phason-related excitations are also observed in complex crystal structures known as approximants. This article briefly reviews the concepts of phasons with focus on current issues in their regard. We examine both continuum (hydrodynamic) and discrete (tiling) descriptions of phasons and review the state of experimental validation of the basic theoretical notions.

Keywords: phason; aperiodic materials; approximants; atomic dynamics; defects in solids; disorder; quasicrystals

1. Introduction

Quasicrystals possess excitations known as phasons [1] as an automatic consequence of their spatial quasiperiodicity. Indeed, phasons were first recognized in other quasiperiodic forms of matter such as incommensurate phases, spin and charge density waves, etc. In quasicrystals, phasons appear in a variety of distinct physical forms, all related to special types of atomic displacements, with associated special names. Some of the terms, which will be discussed further in the following, include [2] phason mode, phason-shift, phason-strain, phason-hop, phason-fluctuation, phason-flip, etc. The study of phasons in quasicrystals has grown into a rich field with extensive theory, simulation and experiments as outlined below.

A word on nomenclature is needed. Some researchers [3,4] suggest reserving the term 'phason' exclusively for phason-*modes*. That means patterns of special atomic displacements that vary in space with a precisely defined wavevector. That seems unnecessarily restrictive as it excludes, for example, strain fields created by internal defects, and also misses the essential connection between the phason mode and the localized atomic rearrangements that comprise it. Localized phenomena such as phason-hops, phason-fluctuations and phason-flips can be represented as linear superpositions of phason modes of differing wavevectors in a manner similar to the representation of an atomic vibration as a superposition of phonons. However, care must be taken to distinguish local degrees of freedom that are unrelated to phason modes from those that are so related. A rule of thumb is that motion of atoms is phason-related when it is conveniently described using either the density wave or perpendicular space descriptions outlined below.

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The following section briefly reviews the formal theory of phasons in quasicrystals. Following that we survey the status of experimental and simulation study of phasons. We start with static 'quenched' non-equilibrium strains, then address 'annealed' equilibrium phason-fluctuations, followed by studies of phason dynamics. Our discussion initially focuses on quasicrystals in metal alloys and concludes with a survey of novel hydrodynamic and metamaterial quasicrystals.

2. Formal theory

The phenomenon of a phason is best understood by representing the actual discrete atomistic structure as a superposition of density waves [5] (see Figure 1a)

$$\rho(\mathbf{r}) = \sum_{k=1}^{N} \rho_{\mathbf{G}_k^{\|}} e^{i\mathbf{G}_k^{\|}\cdot\mathbf{r}}, \tag{1}$$

where the set of $\{\mathbf{G}^{\|}\}$ correspond to reciprocal lattice vectors whose dimensionality d matches the dimensionality of the spatial position variable $\mathbf{r}$. This density could represent, for example, the spatially varying electron density. To accurately represent a particular atomistic structure, the density should include rapidly oscillating terms with large $|\mathbf{G}^{\|}|$. We instead adopt a 'coarse-grained' or 'hydrodynamic' [6] perspective, in which only a small set of N distinct wavevectors $\mathbf{G}^{\|}$ is considered, that preserves essential features of the atomistic structure.

The indexing dimension D of a crystal is the minimum number of reciprocal lattice vectors $\mathbf{G}_k^{\|}$ needed to fully generate the reciprocal lattice by linear combinations with integer coefficients, $\mathbf{G}^{\|} = h_1\mathbf{G}_1 + h_2\mathbf{G}_2 + \cdots + h_D\mathbf{G}_D$. The integers $(h_1, h_2, \ldots, h_D)$ are the D Miller indices of the Bragg peak at wavevector $\mathbf{G}^{\|}$. For a periodic crystal the indexing dimension $D = d$.

Incommensurate phases differ from periodic crystals in that additional lattice vectors are needed in order to index all the diffraction peaks, requiring $D > d$ [7,8]. Ordinary incommensurate structures may have a single extra lattice vector such as $\mathbf{G}_{d+1}^{\|} = f\mathbf{G}_1^{\|}$ where f is continuously variable (and usually irrational) representing the incommensurability of two intrinsic lengths, resulting in $D = \mathrm{d} + 1$. Multiple incommensurabilities are also possible. Quasicrystals differ from ordinary incommensurate crystals in that the reciprocal lattice vectors belong to a fixed set with the incommensurability constrained by symmetry. For icosahedral quasicrystals it is common to use either six non-collinear vertices of an icosahedron [9], or an alternate set related to icosahedron edges [10], with $D = 6$ in each case.

Overcomplete sets with $N > D$ are sometimes used to better represent an underlying symmetry (e.g. $N = 4$ Miller indices for hexagonal crystals in dimension $d = 3$) but in this case there exist integer combinations of $\{\mathbf{G}_k^{\|}, k = 1, \ldots, N\}$ that sum to the zero vector, causing the Miller indexing to become non-unique. Similarly, a five-fold star $(\mathbf{G}_k^{\|} = (\cos 2\pi k/5, \sin 2\pi k/5), k = 1, \ldots, 5)$ is commonly used for pentagonal or decagonal symmetry resulting in $N = 5$ even though $D = 4$.

In the hydrodynamic view, phasons and phonons are closely related. Consider the pattern described by Equation (1). Multiplying each coefficient $\rho_{\mathbf{G}_k^{\|}}$ by a phase factor $e^{i\mathbf{G}_k^{\|}\cdot\mathbf{u}}$ is equivalent to shifting the pattern $\rho(\mathbf{r})$ to $\rho(\mathbf{r}+\mathbf{u})$, that is, translating the pattern by $-\mathbf{u}$. Owing to translation symmetry of space, such a shift costs no energy. The pattern can be

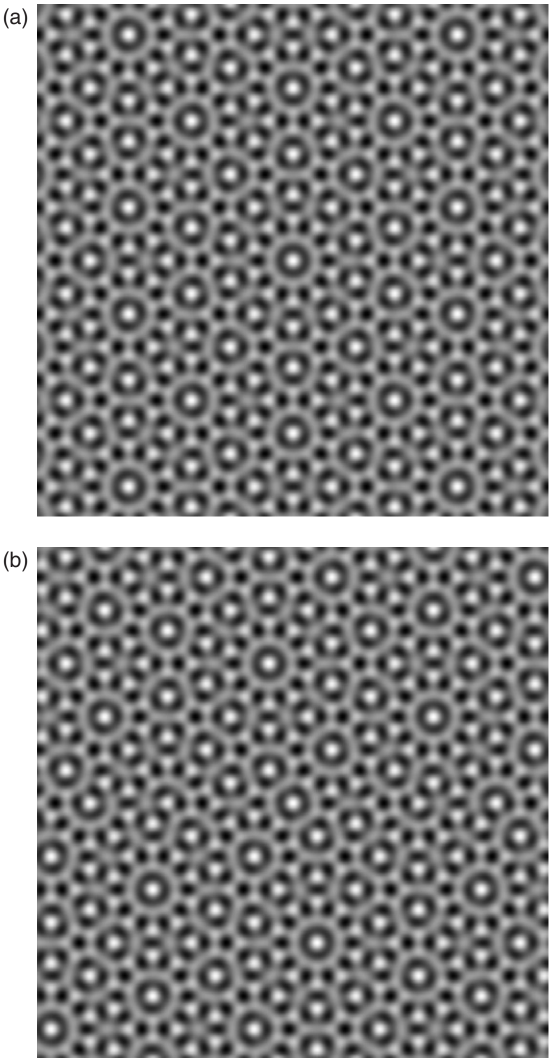


Figure 1. (a) Strain-free quasicrystalline density wave according to Equation (1). (b) Quenched phason strain (view at glancing angles to see zig-zags in rows of contrast).

shifted by different amounts in different regions of space by allowing $\mathbf{u}(\mathbf{r})$ to be a slowly varying function of position. Now the shift does raise the energy, by an amount governed by the spatial gradients $\nabla\mathbf{u}(\mathbf{r})$ (phonon strains) and by the elastic constants of the material under study, such as bulk and shear modulus. If $\mathbf{u}(\mathbf{r})$ varies periodically, say as $e^{i\mathbf{q}\cdot\mathbf{r}}$, the energy varies as $|\mathbf{q}|^2$ and we term the distortion a phonon mode of wavevector $\mathbf{q}$. Since the energy vanishes in the limit of long wavelength, phonon excitations are termed 'Goldstone modes'.

Now instead multiply each coefficient by $e^{i\mathbf{G}_k^{\perp}\cdot\mathbf{w}}$ where the set $\mathbf{G}_k^{\perp}$ corresponds to the original $\mathbf{G}_k$ permuted in a special way governed by the rotational symmetry of the quasicrystal under study (e.g. $\mathbf{G}_k^{\perp} = \mathbf{G}^{\parallel}_{(3k \bmod 5)}$ for pentagonal or decagonal symmetry).

If $\mathbf{w}$ is uniform throughout space, we term such a transformation a 'phason shift'. It can be shown that a phason shift is equivalent to a rigid spatial translation $\mathbf{u}$, but the translation $\mathbf{u}$ depends on $\mathbf{w}$ in such a way that it is usually infinite in magnitude. Again, spatial homogeneity assures the energy cost vanishes, so this phason shift is another Goldstone mode. As was the case for the phonon shift, we can introduce a slow spatial variation into $\mathbf{w}(\mathbf{r})$ in which case the energy cost grows by an amount governed by the spatial gradients $\nabla\mathbf{w}(\mathbf{r})$ (phason strains) and corresponding phason elastic constants. If $\mathbf{w}(\mathbf{r})$ varies periodically, say as $e^{i\mathbf{q}\cdot\mathbf{r}}$, the energy varies as $|\mathbf{q}|^2$ and we term the distortion a phason mode of wavevector $\mathbf{q}$. Notably, a coupling exists between phonon and phason degrees of freedom.

The extra dimensions needed for indexing lie at the origin of the popular higher-dimensional space representations of quasicrystals and tilings [11–13]. For example, taking tile edges as vectors belonging to $\mathbf{G}_k$, a tiling vertex located at $\mathbf{r} = \sum N_k \mathbf{G}_k$ may be represented as the point $\mathbf{z}(\mathbf{r}) = (N_1, N_2, \ldots, N_D)$ in the D-dimensional hypercubic lattice and the N_k can be evaluated simply by following the edges connecting $\mathbf{r}$ to the origin. Connectivity of the tiling implies that the D-dimensional coordinates lie on a continuous hypersurface of dimension $d < D$. Idealized atomic positions in real $d = 3$-dimensional space are found by intersecting real space with a set of 'atomic surfaces' located at D-dimensional lattice positions [14].

3. Quenched phason strain

Unlike phonons, which correspond to small atomic displacements and are easily relaxed, phasons involve discrete atomic displacements (see below) and are presumed to be difficult to relax. This allows observation of phason strain as a semi-permanent property of a sample, though it can be annealed out through careful heat treatment in some cases.

Strong phason strain was observed in many early quasicrystal-forming compounds. The experimental signatures of phason strain include: shifting of selected-area electron diffraction peak positions in an unusual pattern of alternating signs [15]; zig-zags in rows of contrast in high resolution electron microscope images [16]; broadening of X-ray diffraction peaks [17]. Figure 1(b) illustrates a density wave pattern in which the strain field $\mathbf{w}(\mathbf{r}) = \hat{x}\tanh(y/10)$ has been imposed causing zig-zags in rows of contrast similar to those seen in electron microscope images.

Phason strain occurs in the vicinity of many defects, notably dislocations. Dislocations are characterized by their Burgers vector $\mathbf{b}$, a topological property that can be calculated by counting the number of forward or backward steps taken in each independent direction on a path encircling the dislocation. Consequently, the Burgers vector is an integer combination of translation vectors of the lattice. In quasicrystals the Burgers vector has dimension D, with both parallel and perpendicular space components $\mathbf{b}^{\parallel}$ and $\mathbf{b}^{\perp}$ [18]. The energy of a dislocation is a sum of parallel and perpendicular space contributions, each term being quadratic in the parallel and perpendicular parts of the Burgers vector and proportional to corresponding elastic constants. Interestingly, mechanical deformation of the quasicrystal tends to drive the population of dislocations towards Burgers vectors with small $\mathbf{b}^{\parallel}$ and large $\mathbf{b}^{\perp}$ components, suggesting that the phason elastic constants are small compared to the ordinary phonon elastic constants (i.e. bulk and shear moduli) [19].

Figure 2. Predicted diffuse scattering pattern along icosahedral two-fold axis [26]. Range of plot is ±4 Å^{-1} in horizontal direction and 0-4 Å^{-1} in vertical direction.

4. Annealed phason strain

Since phonon and phason strains are gradients of the displacement fields **u** and **w**, and the elastic energy is quadratic in the strains [5], the elastic energy can be written in reciprocal space as

$$F[\mathbf{u}(\mathbf{r}),\mathbf{w}(\mathbf{r})] = \int d\mathbf{q} K_{ij}(\mathbf{q}) v_i(\mathbf{q}) v_j(\mathbf{q}) \tag{2}$$

where the vector $\mathbf{v} = (\mathbf{u}, \mathbf{w})$ has D components, the first d of which are usual spatial variables corresponding to phonons and the remaining $D - d$ correspond to phasons. The matrix $K_{ij}(\mathbf{q})$ is quadratic in $\mathbf{q}$ and has a complicated orientational dependence. From equipartition we find that the mean-square fluctuations in $\mathbf{v}$ are

$$\langle v_i(\mathbf{q}) v_j(-\mathbf{q})\rangle = k_B T K_{ij}^{-1}. \tag{3}$$

Thus we expect phason fields to fluctuate in thermal equilibrium, as is the case for phonons. One of the expected consequences of phason fluctuations is a Debye–Waller factor reducing diffraction peak intensities according to

$$e^{-2W} = e^{-G_i G_j \langle v_i v_j \rangle} \tag{4}$$

where $\mathbf{G} = (\mathbf{G}^{\parallel}, \mathbf{G}^{\perp})$. Such Debye–Waller effects related to the $\mathbf{G}^{\perp}$ values have been observed [20]. A second consequence of the fluctuations is to create patterns of diffuse scattering whose orientational behaviour reflects the complex orientational dependence of $K(\mathbf{q})$ [21–23]. These patterns have been observed in many important quasicrystal-forming compounds [24–27].

Both the Debye–Waller and diffuse scattering effects depend on composition and temperature because the elastic constant matrix $K(\mathbf{q})$ should vary according to these physical parameters. In particular it can become singular as phase transitions are approached and the material loses elastic stability (a necessary criterion for thermodynamic stability). Indeed, enhanced Debye–Waller and diffuse scattering effects arise in such circumstances. A curious feature is that the stable quasicrystal phase appears to be the high temperature phase, which indicates the phason elastic constants contain terms proportional to temperature and the state is stabilized by configurational entropy [22,28].

5. Dynamic phason strain

Assuming the elastic free energy given in Equation (2), the hydrodynamic theory of quasicrystals [6] predicts a relaxation-type dynamics for phasons described by

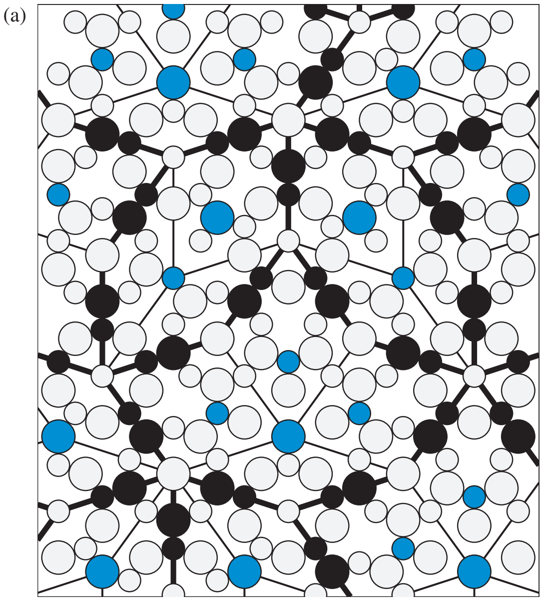
$$\frac{\partial \mathbf{w}(\mathbf{r}, t)}{\partial t} = -\Gamma \frac{\delta F}{\delta \mathbf{w}(\mathbf{r}, t)} \tag{5}$$

where Γ_w is a kinetic coefficient. Fourier transforming this equation yields a solution decaying exponentially in time with an exponential decay rate $\omega(\mathbf{q}) = \Gamma_\mathrm{w} K_\mathrm{w}(\mathbf{q})$ (here K_w represents the phason part of $K(\mathbf{q})$ and we neglect phonon–phason coupling). Recalling that $K_w(\mathbf{q}) \sim q^2$, we see that short wavelength, high q modes decay quickly, while long wavelength, small q modes decay slowly in time. Experiments on dynamics of X-ray speckle patterns in icosahedral AlPdMn [29] observe the expected q-dependent decay with decay times ranging from 20–70 seconds over the observed q range of $1.5\text{–}6 \times 10^7\,\mathrm{m}^{-1}$ at temperature $T = 6504°\mathrm{C}$.

Dynamics of dislocations provide further insight into the dynamics of phason strain fields. Long range strain fields surround the dislocation line with phonon and phason components proportional to the magnitude of the parallel and perpendicular components of the Burgers vector. Motion of the dislocation line demands that the strain fields also must move. While the phonon strain field can relax quickly through small atomic displacements, relaxation of the phason strain field requires discrete and correlated atomic jumps that may be difficult to achieve. Consequently the dislocation leaves behind a trail of phason flips in the form of a planar fault known as a 'phason wall' or 'phason trail' [30]. In i-AlPdMn the phason field of this defect decays over temperature-dependent time scales ranging from 1 to 10^5 seconds, obeying an Arrhenius temperature dependence with activation energy of 4eV [31]. The temperature and time scales in phason wall relaxation are consistent with those seen in X-ray speckle patterns [29].

Since the D-dimensional coordinates are integers, local deformations of the phason coordinate $\mathbf{w}$ cause discontinuous jumps in the hypercubic lattice position $\mathbf{z}(\mathbf{r})$. This, in turn, has the effect of flipping the configuration of tiles in the vicinity of the point $\mathbf{r}$. The tiling shown in Figure 3 consists of fat and thin Penrose rhombi and has been decorated with atoms as appropriate for decagonal AlNiCo [32]. An alternate description in terms of hexagons, boats and stars is indicated using heavy edges. In the original tiling (left) the boat is upright, while it is overturned in the flipped tiling (right). The central region of the figures illustrate the famous decagonal cluster with 20 Å diameter and symmetry-broken interior. The particular tile flip illustrated here leaves the entire cluster intact, but rotates the symmetry-breaking feature in the interior. Other flips (not shown) are able to create or destroy the clusters.

Notice that the flip has interchanged the positions of two fat and one thin Penrose rhombus, leaving all other tiles unaffected. Since the tile flip is localized, only a few



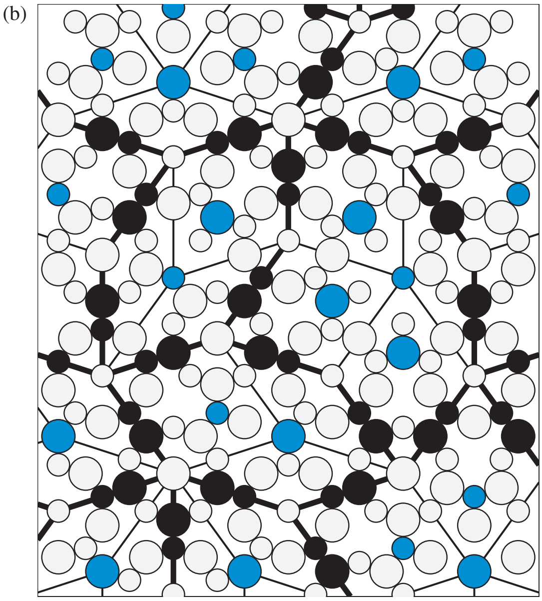


Figure 3. HBS tilings of d-AlCoNi (a) boat upright (b) boat flipped. Atomic positions are indicated as Al = white, Co = blue, Ni = black. Large/small circles indicate vertical position. Tile edge length is 6.5 Å.

atomic positions change, even though the result is a rotation of the entire 20 Å cluster. Further, the atoms in the vicinity of the tile flip are in highly symmetric positions so that no change in interatomic distances occurs out to a considerable distance, and the energies before and after the flip are nearly equal. Counting the possible orientations of the cluster center and the symmetry-breaking within the hexagon tiles yields a total of 40 distinct configurations simply within the 20 Å cluster. The combination of near-degeneracy with high multiplicity motivates the proposal of entropic stabilization of the quasicrystal state [28,33] in the random tiling scenario [11,34].

Creation and destruction of 20 Å clusters has been directly observed in high resolution electron microscopy performed *in situ* at elevated temperature on decagonal d-AlCuCo [35,36]. The structure of d-AlCuCo is expected [37] to resemble that shown in Figure 3 for d-AlNiCo, and the observed cluster dynamics is believed to be related to flips such as those illustrated here. The flips occur more frequently in thin regions of the sample (on a time scale of order 1 second) than in thick regions. Presumably it takes longer for the flip to propagate through a thick region of the sample than for a thin region. Recall that HRTEM observed projected structures, so if a flip extends only part way through the sample, it might not be observed.

The smallest scale atomic motion that can be considered as phason-related is the hopping of a single atom between ideal positions. In Figure 3 this corresponds to the reversal of Penrose rhombi comprising a Hexagon tile, and involves the displacement of a single Al atom by 2.2 Å. In models with higher densities of ideal sites the minimum displacements can be even smaller.

Single- and multiple-atom jumps have been observed experimentally in several contexts. The first direct reports came from quasielastic neutron scattering experiments on i-AlCuFe [38] and i-AlPdMn [39]. These experiments reported hopping over length scales around 4 Å and time scales of hundreds of picoseconds ($\hbar/E$ with E in the range 2–200 μeV) [40]. Less direct evidence comes from the observation of enhanced Debye–Waller factors in electron microscopy studies of d-AlNiCo [41], interpreted as 0.95 Å jumps between ideal sites. Significantly, these highly mobile atomic positions occur on atoms projected from the edges of higher dimensional atomic surfaces, where phason hopping is expected to be most prevalent. Unfortunately the time scale for hopping is not resolved in this experiment.

Since localized phason hops involve displacements of atoms a natural question is the role of phason in long-range atomic diffusion. Phason-assisted diffusion was first proposed by Kalugin and Katz [42] and has been observed in computer simulation [43]. Experimental NMR studies initially found only short-ranged phason assisted diffusion [44] and this occurred on kilohertz time scales at rather low temperatures $T \leq 160$ K. Later studies found evidence for percolating diffusion pathways [45], still at low temperatures and on fast time scales. Tracer diffusion studies of ^{103}Pd and ^{195}Au in i-AlPdMn observe conventional vacancy controlled diffusion at high temperature while diffusion below $T = 450°$C is tentatively identified as phason-assisted [46].

6. Phason-like behaviour in approximants

Approximants are periodic crystals with large unit cells that contain quasicrystal-like structure within the unit cell. They can be described as tilings of space using the basic tile building blocks found in quasicrystals of similar chemical composition, but the tiles are arranged in a spatially periodic manner rather than quasiperiodically.

Owing to the crystal periodicity the phason loses its status as a Goldstone mode arising from a continuous symmetry. Still, local manifestations of phasons such as localized tile flips and atomic hopping may occur as a type of atomic disorder in the crystal. Such localized atomic jumps actually appear in an order-disorder transition in the 1/1 cubic approximants of Zn- and Cd-based icosahedral phases [47]. This transition occurs at a surprisingly low temperature of $T = 80\,\mathrm{K}$ indicating a very nearly perfect degeneracy of differing configurations and low barriers separating them. The energy differences are probably so low because the transition occurs in the orientations of symmetry-breaking clusters buried near the center of a large highly symmetric cluster, and the coupling between centers of adjacent clusters is weak [48].

Since approximants are periodic crystals they have ordinary dislocations with Burgers vectors containing only a parallel space component $\mathbf{b}^{\parallel}$. However, the large unit cells of the approximants create large strain energies even for the smallest possible Burgers vector dislocations. Dislocation energy can be reduced by splitting into partials, each carrying a portion of the full Burgers vector $\mathbf{b}^{\parallel}$ but acquiring at the same time a phason component $\mathbf{b}^{\perp}$ [49]. The dislocation core spreads into a broad region known as a metadislocation that bounds on one edge a wall of stacking faults. The stacking faults consist of phason-flipped regions of the tiling corresponding to the approximant phase. Interestingly, the flipped region can yield a new periodic tiling corresponding to other known approximant crystal structures, such as conversion of the orthorhombic $Al_{13}Co_4$ structure into a monoclinic variant [50].

7. Phasons in metamaterials

Metamaterials are an emerging area of quasicrystal research. In these materials patterns emerge at length scales greater than atomic dimensions and can be observed with optical microscopy and in some cases even with the naked eye. Quasicrystalline patterns have been observed in such diverse contexts as Faraday wave hydrodynamics [51–53], nonlinear photonic structures [54,55] and soft-matter systems such as lyotropic liquid crystals and ABC star polymers [56–58]. The origins of stability of quasicrystals in these metamaterials may differ from the origin in metal alloys [59].

Phason strain has been directly observed in Faraday wave experiments perturbed with an additional frequency of forcing [60]. In this case the strain achieves a conversion of a periodic triangular pattern into a hexagonal pattern, in a manner reminiscent of the conversion between alternative crystal structures by a metadislocation. Hydrodynamic phason dynamics can also be simulated using the Lifshitz–Petrich equation with the phason field arising from a dislocation [61].

Phasons have been deliberately introduced in nonlinear photonic materials [55]. Their relaxation times are slower than phonon relaxation times, and strain-relaxing tile flips have been directly visualized.

Observed patterns in both dendritic liquid crystals and ABC star polymers correspond to tilings of triangles and squares. The dodecagonal quasicrystal state of this tiling is known to be a maximal entropy intermediate between a six-fold symmetric triangular tiling and a four-fold symmetric square tiling [62]. For dendritic liquid crystals, intrinsic phason disorder is apparent in the observed electron diffraction pattern [56] which exhibits peak shifts and broadening. The dodecagonal quasicrystal phase observed in ABC star polymers has the appearance of a random tiling with strong phason fluctuations [58].

8. Future outlook

Study of phasons and phason-related phenomena in quasicrystals and their approximants has a rich and promising future [2]. It is heartening that qualitative agreement is found between differing experimental probes, especially the consistent time and energy scales found in i-AlPdMn where equilibrium fluctuations observed by X-ray speckle patterns [29] qualitatively match with non-equilibrium relaxation of phason wall defects created by dislocation line motion [31]. Still unresolved is the precise link of these large-scale and slowly evolving phasons to the highly localized single- or few-atom hops such as those observed in quasielastic neutron scattering [40] and electron microscopy [36,41]. Presumably the large scale behaviour can be explained through a series of distinct small scale events, but this has not yet been achieved.

Also lacking is a precise quantitative link between equilibrium phason fluctuations and thermodynamic stability. It is now possible to estimate the energies of some phason flips using first-principles calculations [63] and predict their contribution to heat capacity and other thermodynamic properties which can be compared with experiment. Ultimately the phason elastic constants can be estimated and compared with experimental diffuse scattering data [24], though this will require simultaneous improvements in total energy calculation and structure modelling. Pursuing the influence of phasons on thermodynamic stability can help resolve the long-standing question of energetic vs. entropic stabilization [64].

The study of phasons in metamaterials offers great possibilities, because phason can be deliberately introduced in controlled fashion [55,61] and directly observed. Although the behaviours of phasons in these novel quasicrystalline materials may not shed direct light on phasons in metal alloy quasicrystals, similar concepts apply and similar questions (e.g. elasticity and hydrodynamics) may be asked.

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