Phason elastic constants of a binary tiling quasicrystal

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Abstract

For a two-dimensional binary tiling model quasicrystal, the full set of (zero temperature) elastic constants is determined. It is found that the elastic energy is a perfect quadratic form in the phonon and phason strains. One of the phason elastic constants turns out to be negative, implying that the quasicrystal is only metastable at zero temperature.

Keywords: Quasicrystals; Elasticity

1. Introduction

Elastic deformations of quasicrystals can be either of phonon type, or of phason type. While the former occur also for ordinary crystals, the latter exist only for quasicrystals and incommensurately modulated crystals (from where the name has been borrowed). There is an effective elasticity theory also for the phasonic degrees of freedom. In fact, phasons contribute in different ways to the free energy of the system. On one hand, there is an energetic term. A phasonic deformation changes the frequences of the different local environments in the structure, and thus changes the energy of the system. On the other hand, *local* phasonic distortions are of relatively low energy, and are easily accessible at high temperature. The number of possible such excitations depends on the *average* phason strain. The lower the average phason strain, the higher the number of possible excitations. This mechanism is at the heart of the random tiling model [1]. It leads to an entropic part of the free energy, and thus to a contribution to the effective phason elastic constants.

The first estimates of the entropic part of the phason elastic constants were done on purely geometric random tiling models, either by Monte-Carlo simulations [2, 3, 4] or by transfer matrix methods [5]. Density functional theories [6, 7] were able to determine the full set of elastic constants, but depended on many unknown other system parameters. In real experiments the anisotropic diffuse scattering close to Bragg peaks was used to determine first the ratio of the two phason elastic constants [8], and recently also their absolute values [9]. Naturally, these experimental values included both energetic and entropic terms. To find the phason-phonon coupling constant, Zhu and Henley [10] relaxed a phason strained icosahedral model quasicrystal and measured the spontaneous phonon strain that developed in the relaxation. The method of Zhu and Henley works at zero temperature. It can be used also for the determination of the energetic part of the phason elastic constants. Unfortunately, the entropic part is inaccessible with this method. It is expected that the entropic contribution to the phason-phonon coupling is not important.

In the present work, we use methods similar to [10], working with a simple decagonal model quasicrystal in two dimensions. We determine the full set of elastic constants of this system, with the proviso that only the energetic parts in the phason elastic constants are included. The remainder of the paper is organized as follows. In section 2, our model system is introduced, and in section 3 we give a short review of elasticity theory of decagonal quasicrystals. Our simulations are described in section 4, where we also present the measured elastic constants. Finally, these results are discussed in section 5, and put into a broader context.

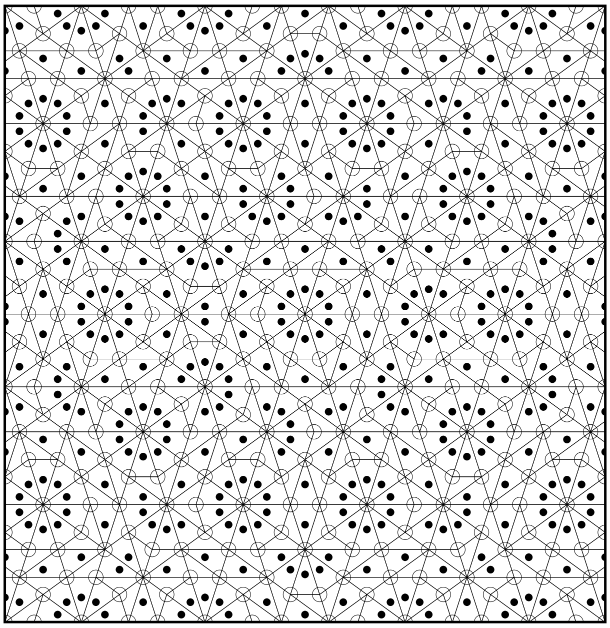

Fig. 1. Mikulla-Roth binary tiling as a decoration of the Tübingen triangle tiling.

2. The quasicrystal model

The atomic configuration of our model is a so-called binary tiling structure [11, 12]. There are many different variants of binary tiling structures, which can be quasiperiodic or of random tiling type [12]. We choose here the (quasiperiodic) binary tiling of Mikulla and Roth (Fig. 1), first mentioned in [13]. Its construction based on the Tübingen triangle tiling [14] is described in [12]. The advantage of this model is that it is very easy to construct many different periodic approximants of the Tübingen triangle tiling, which can be decorated in the same way. This is important for our simulations (see section 4).

If all bonds have their ideal length, and the interactions are of sufficiently short range so that only first neighbours are seen, then all binary tilings of a given stoichiometry are energetically degenerate [11]. There is a trivial dependence on stoichiometry, however: the higher the density of small atoms, the higher is the density of interacting bonds. In order to be able to distinguish between different local environments, we have to choose interaction potentials with a longer range. For simplicity, we have chosen Lennard-Jones potentials (although it is unlikely that these will have a quasiperiodic ground state):

$$\phi_{ij}(r) = \epsilon_{ij} \left((\frac{\sigma_{ij}}{r})^{12} - 2\,(\frac{\sigma_{ij}}{r})^{6} \right) \qquad (1)$$

where r is the distance of the two atoms and i, j are the atom types. The values of the parameters are

$$\begin{aligned} \epsilon_{AA} &= 1 & \epsilon_{AB} &= 2 & \epsilon_{BB} &= 1 \\ \sigma_{AA} &= 1.176 & \sigma_{AB} &= 1 & \sigma_{BB} &= 0.6180 \end{aligned} \qquad (2)$$

They are chosen such that bonds beween atoms of different type are preferred, which makes phase separation unfavourable. For performance reasons the potentials are cut off at a radius of $R = 7$. In addition, the potentials are shifted so that they vanish at the cutoff radius R:

$$\tilde{\phi}_{ij}(r) = \phi_{ij}(r) - \phi_{ij}(R) \qquad (r < R) \qquad (3)$$

3. Phason-phonon elasticity theory

The elastic energy density $E(\varepsilon_{ij}, \chi_{ij})$ of a quasicrystal is a function of both the usual phonon strain $\varepsilon_{ij} = (\partial u_i^{\|}/\partial x_j^{\|} + \partial u_j^{\|}/\partial x_i^{\|})/2$ and the quasicrystal-specific phason strain $\chi_{ij} = \partial u_i^{\perp}/\partial x_j^{\|}$. Decomposing the phonon strain with respect to irreducible representations of the D_{10} symmetry of the decagonal quasicrystal, we get two phonon modes, the bulk deformation $\varepsilon^{(1)}$ and a (two-component) shear deformation $\boldsymbol{\varepsilon}^{(6)}$. A similar decomposition of the phason strain yields two modes $\boldsymbol{\chi}^{(6)}$ and $\boldsymbol{\chi}^{(8)}$ transforming under different representations, with two components each (the upper index labels the corresponding irreducible representation of D_{10}). For the free elastic energy density, this results in the following decomposition into symmetry invariant terms:

$$\begin{aligned} E =& E_{\text{phonon}} + E_{\text{phason}} + E_{\text{coupl.}} \qquad (4) \\ E_{\text{phonon}} =& \frac{1}{2}\lambda_3(\varepsilon^{(1)})^2 + \frac{1}{2}\lambda_5[(\varepsilon_1^{(6)})^2 + (\varepsilon_2^{(6)})^2] \\ E_{\text{phason}} =& \quad \frac{1}{2}\lambda_7[(\chi_1^{(6)})^2 + (\chi_2^{(6)})^2] \\ & + \frac{1}{2}\lambda_9[(\chi_1^{(8)})^2 + (\chi_2^{(8)})^2] \\ E_{\text{coupl.}} =& \lambda_6[\varepsilon_1^{(6)}\chi_1^{(6)} + \varepsilon_2^{(6)}\chi_2^{(6)}] \end{aligned}$$

where $\lambda_3/2$ is the bulk modulus, $\lambda_5/2$ the shear modulus, λ_7 and λ_9 are two phason elastic constants, and λ_6 is the coupling constant between the shear mode and the first phason mode.

4. Simulations and results

In our simulations, we measure the minimum of the potential energy as a function of phonon and phason strain, using our molecular dynamics program IMD [15]. Up to a constant, this corresponds to the zero temperature elastic free energy. In order to avoid surface effects, we use periodic approximants. For each approximant, the average phason strain has a fixed value, which is given by the geometry of the approximant (see Table 1). We can thus vary the phason strain only by using many different approximants, whereas the phonon strain can be varied by applying a linear transformation to the atom coordinates. For each approximant, the phonon strain with minimal energy is determined first. In a second

τ_x	τ_y	$\chi_1^{(6)}$	$\chi_1^{(8)}$	A/B
123/80	58/37	-0.03265	$-1.9 \cdot 10^{-7}$	0.80865
63/40	218/137	-0.01715	$-1.0 \cdot 10^{-7}$	0.80892
78/49	181/113	-0.01036	$1.6 \cdot 10^{-7}$	0.80898
269/168	127/79	-0.00663	$-2.7 \cdot 10^{-7}$	0.80900
177/110	129/80	-0.00351	$-5.1 \cdot 10^{-7}$	0.80901
316/195	149/92	0.00097	$0.4 \cdot 10^{-7}$	0.80902
127/78	294/181	0.00396	$-0.6 \cdot 10^{-7}$	0.80901
289/177	96/59	0.00572	$-3.7 \cdot 10^{-7}$	0.80901
79/48	121/74	0.01073	$-4.4 \cdot 10^{-7}$	0.80898
168/101	79/48	0.01737	$7.1 \cdot 10^{-7}$	0.80891
49/29	113/68	0.02713	$-4.2 \cdot 10^{-7}$	0.80876
74/47	79/48	$-7.1 \cdot 10^{-7}$	-0.01737	0.80912
144/91	105/64	$-10.7 \cdot 10^{-7}$	-0.01415	0.80909
167/105	157/96	$14.0 \cdot 10^{-7}$	-0.01091	0.80906
67/42	111/68	$-18.7 \cdot 10^{-7}$	-0.00900	0.80904
85/53	314/193	$11.6 \cdot 10^{-7}$	-0.00561	0.80903
140/87	138/85	$-7.2 \cdot 10^{-7}$	-0.00347	0.80902
92/57	316/195	$0.6 \cdot 10^{-7}$	-0.00157	0.80902
295/182	139/86	$-0.5 \cdot 10^{-7}$	0.00111	0.80902
294/181	205/127	$0.4 \cdot 10^{-7}$	0.00245	0.80902
119/73	182/113	$1.9 \cdot 10^{-7}$	0.00471	0.80902
113/69	53/33	$-3.1 \cdot 10^{-7}$	0.00761	0.80904
71/43	163/102	$2.0 \cdot 10^{-7}$	0.01276	0.80907
113/68	78/49	$2.6 \cdot 10^{-7}$	0.01677	0.80911

Table 1. Approximants used in the simulations. Listed are the two rational values approximating the golden mean in x and y direction, the two phason strain components, and the stoichiometric ratio between big and small atoms.

step, the phonon strain is then varied around this value. While varying the phonon strain, the structure is permanently kept in its potential minimum. All that can easily be done with the infrastructure built into IMD [15].

In order to facilitate the interpretation of the simulation results, we have used approximants which have nearly pure mode phason strains, transforming either by $\Gamma^{(6)}$ or $\Gamma^{(8)}$ (see Table 1). The bulk and shear moduli, $\lambda_3/2$ and $\lambda_5/2$, are easy to determine by applying the corresponding phonon strains to the different approximants (see Fig. 2). The phonon elastic constants correspond to the curvature of the parabolas at their minimum. They do not depend on the choice of the approximant. We also see that the minimum of the potential energy is realized for a non-zero shear deformation (see Fig. 2(b)), which is due to phason-phonon coupling. The position of the minimum depends only on the phason strain $\chi_1^{(6)}$, and not on $\chi_1^{(8)}$, which is in agreement with the symmetry analysis in section 3. This can be seen also in Fig. 3, where the elastic energy is plotted against the two types of phason strain. The different curves correspond to different shear deformations. We see again, that the optimal $\chi_1^{(6)}$ phason strain depends on the given shear deformation, but not the optimal $\chi_1^{(8)}$ phason strain. The curvatures at the extrema of the parabolas in Fig. 3 correspond to the two phason elastic constants λ_7 and λ_9. We note that λ_7 is negative, which means that the quasicrystal is only locally

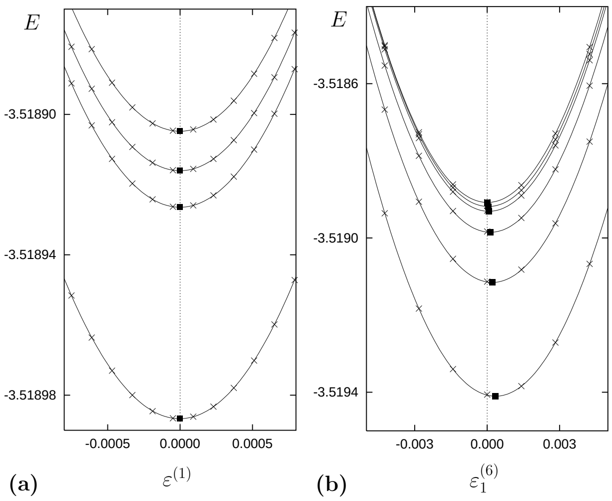

Fig. 2. Elastic energy density against bulk and shear deformation for different phasonic deformations $\chi_1^{(6)}$.

stable (metastable). The phason-phonon coupling is best evaluated by plotting the extremum positions of Fig. 3(a) against the shear deformation (Fig. 4). This reveals a linear relationship, whose proportionality constant is $-\lambda_6/\lambda_7$, from which the phason-phonon coupling constant λ_6 can be derived.

Summarizing, we obtain the following values for the elastic constants of our model system:

$$\begin{array}{rll} \text{phonon:} & \lambda_3 = 125 & \lambda_5 = 45.1 \\ \text{phason:} & \lambda_7 = -1.35 & \lambda_9 = 0.4 \\ \text{coupling:} & \lambda_6 = -0.57 & \end{array}$$

The dimension of all values is $[\epsilon_{AB}/\sigma_{AB}^2]$.

5. Discussion

We have determined the full set of (zero temperature) elastic constants of a two-dimensional binary tiling model quasicrystal. One of the two phason elastic constants turned out to be negative, which means that this quasicrystal is only metastable at zero temperature.

The most important contribution to the phasonic elastic energy is due to the stoichiometry, which varies with phason strain. In fact, the concentrations of large and small atoms are a function of phason strain, and the potential energy density strongly depends on these concentrations and thus on the phason strain. The higher the concentration of small atoms, the higher is the density of interactiong bonds. While a phason strain of type $\boldsymbol{\chi}^{(6)}$ increases the concentration of small atoms, a phason strain of type $\boldsymbol{\chi}^{(8)}$ decreases it. It is perhaps problematic to compare the energies of structures with

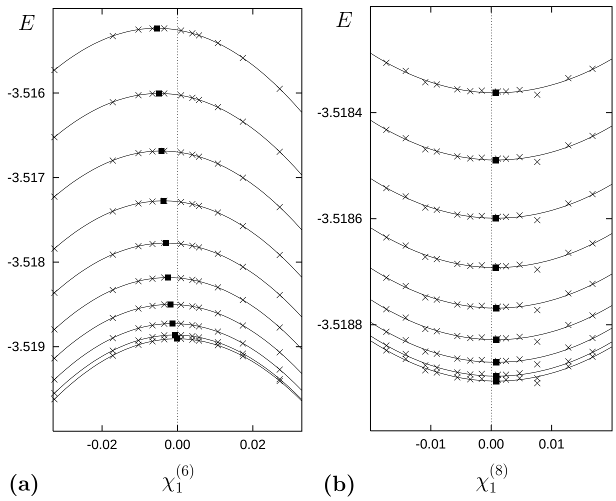

Fig. 3. Elastic energy density as a function of the phasonic deformations $\chi_1^{(6)}$ and $\chi_1^{(8)}$, for several different shear deformations $\varepsilon_1^{(6)}$.

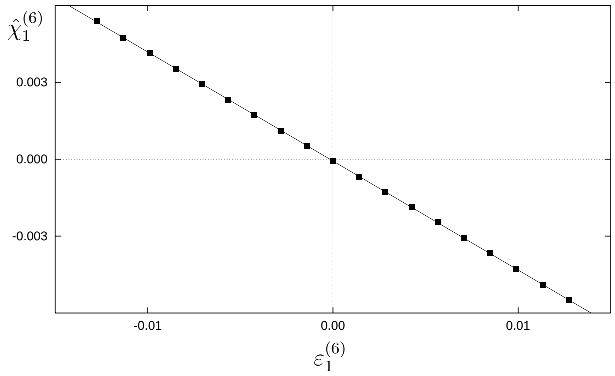

Fig. 4. Extremum positions $\hat{\chi}_1^{(6)}$ from Fig. 3(a) as a function of the shear deformation $\varepsilon_1^{(6)}$, showing the linear behaviour of the phason-phonon coupling.

different stoichiometries, without taking the chemical potentials into account. If the phason strain satisfies $|\boldsymbol{\chi}^{(6)}| = |\boldsymbol{\chi}^{(8)}|$, the stoichiometry remains invariant, but even in those directions the quasicrystal is unstable, because the magnitude of λ_7 is larger than that of λ_9. It should also be emphasized that at higher temperatures, there are also entropic contributions to the phason elastic constants, which might well dominate over the energetic part.

The phason-phonon coupling constant, on the other hand, should not be influenced by these problems. Its determination does not require a comparison of absolute energy densities of structures with different stoichiometries. Also, the phason-phonon coupling should not be influenced by entropic contributions at higher temperatures. It measures the spontaneous shear deformation in reaction to an imposed phason strain. This should not depend on the origin of the phason strain, entropic or energetic.

The elastic energy density turned out to be a perfect quadratic form in the phonon and phason strains. It is common folklore [1, 16] that an energetically stabilized quasicrystal should have an energy which varies linearly with phason strain. These ideas are based on tiling models with simple energetics, where only defects, such as violations of matching rules, are taken into account. These defects correspond to local environments not present in the perfect structure. Even in our case of a quasicrystal which is not stable at zero temperature, there are defects (discontinuities in lattice planes) whose density varies linearly with phason strain. There are, however, not only these defects, but also variations in the densities of those local environments which occur in the perfect quasicrystal. These densities vary quadratically with phason strain. We suspect that these latter contributions are far more important and can overshadow a possible linear part.

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