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Non-linear generalized elasticity of icosahedral quasicrystals

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Abstract

Quasicrystals can carry, in addition to the classical phonon displacement field, a phason displacement field, which requires a generalized theory of elasticity. In this paper, the third-order strain invariants (including phason strain) of icosahedral quasicrystals are determined. They are connected with 20 independent third-order elastic constants. By means of non-linear elasticity, phason strains with icosahedral irreducible Γ^4 -symmetry can be obtained by phonon stress, which is impossible in linear elasticity.

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1. Introduction

Apart from the ordinary phonon degrees of freedom, quasicrystals (QC) have phason degrees of freedom, referring to relative shifts of the constituent density waves [1, 2]. Therefore, and due to their lack of translational order, quasicrystalline structures are usually constructed as an irrational cut of a decorated hyperspace structure by physical space E^{\parallel} [3, 4]. The phason degrees of freedom are connected with the displacement field along the orthogonal space E^{\perp} . The generalized elasticity is described in terms of spatially varying phonon and phason displacement fields [1, 2].

Icosahedral QC have three phonon and three phason degrees of freedom and associated components of a displacement field. Linear elastic theory provides five independent second-order elastic constants, two belonging to pure phonon elasticity, two to pure phason elasticity and one to a coupling between phonons and phasons.

Within linear phonon elasticity, icosahedral QC behave essentially like isotropic media [5]. Faithful icosahedral symmetry exists for physical properties described by tensors of rank $N \geq 5$ only [6]. Accordingly, the non-linear elasticity of icosahedral QC is anisotropic [7].

Fundamental research on classical non-linear elasticity was performed many years ago [8, 9]. The authors of [6, 10–13] have already determined the four linearly independent,

icosahedral elastic tensors of rank six, related to third-order phonon elastic invariants. In contrast to this, in the isotropic case one has only three independent third-order phonon elastic invariants, or elastic constants. Ishii [14] has calculated the pure phason third-order icosahedral invariants. The aim of this paper is to generalize the classical non-linear elasticity and to determine all third-order icosahedral elastic invariants, which occur when phason strains are included. The idea leading to this work was to find a possible method to generate phason Γ^4 -strain in icosahedral QC by phonon stress, which is impossible within linear elasticity.

2. Generalized elastic theory of icosahedral QC

According to their icosahedral diffraction pattern, the mass density of icosahedral QC is a sum of density waves indexed by a reciprocal lattice L of icosahedral symmetry: $\rho(\mathbf{x}) = \sum_{\mathbf{k} \in L} \rho_{\mathbf{k}} \exp(i\mathbf{k} \cdot \mathbf{x}) = \sum_{\mathbf{k} \in L} |\rho_{\mathbf{k}}| \exp[i(\mathbf{k} \cdot \mathbf{x} + \phi_{\mathbf{k}})]$. The phases $\phi_{\mathbf{k}}$ of the basis vectors of L are six degrees of freedom [2], parametrized by the phonon and phason displacement fields \mathbf{u} and \mathbf{w} via $\phi_{\mathbf{k}} = \phi_{\mathbf{k},0} - \mathbf{k}^{\parallel} \cdot \mathbf{u} - \mathbf{k}^{\perp} \cdot \mathbf{w}$. Here, $\mathbf{k}^{\parallel} = \mathbf{k}$ and \mathbf{k}^{\perp} are the projections of reciprocal six-dimensional hyperlattice vectors onto E^{\parallel} and E^{\perp} , respectively. The phonon and phason displacement fields \mathbf{u} and \mathbf{w} are the projections of the hyperspace displacement field $\mathbf{u} \oplus \mathbf{w}$ onto E^{\parallel} and E^{\perp} , respectively.

We denote the position of a point in the undistorted QC \mathbf{a} and the corresponding position in the distorted structure \mathbf{x} , where $\mathbf{x} = \mathbf{u} + \mathbf{a}$. In the Lagrangian scheme, all quantities depend on the variable \mathbf{a} [8]. The phonon strain tensor $\boldsymbol{\eta}^u$ has its components of the classical symmetric form

$$\eta_{ij}^u = \frac{1}{2} \left(\frac{\partial u_i}{\partial a_j} + \frac{\partial u_j}{\partial a_i} \right) + \frac{1}{2} \frac{\partial u_k}{\partial a_i} \frac{\partial u_k}{\partial a_j} \quad (1)$$

or, written in terms of the Jacobian $F_{ij} = \frac{\partial x_i}{\partial a_j}$, $\eta_{ij}^u = \frac{1}{2}(F_{ki}F_{kj} - \delta_{ij})$. This strain tensor is free of rigid rotations. The phason displacement gradient $\frac{\partial w}{\partial a}$ splits into a Γ^4 and a Γ^5 part (see (9)), and both are assumed to increase the elastic energy [2, 15]. Therefore, we have a phason strain tensor $\boldsymbol{\eta}^w$ with

$$\eta_{ij}^w = \frac{\partial w_i}{\partial a_j}. \quad (2)$$

In the linear limit $|\frac{\partial u_i}{\partial a_j}| \ll 1$, the components of $\boldsymbol{\eta}^u$ take their well-known shape $\eta_{ij}^u = \frac{1}{2}(\frac{\partial u_i}{\partial a_j} + \frac{\partial u_j}{\partial a_i})$.

The isothermal Helmholtz free energy $F(\boldsymbol{\eta}^u, \boldsymbol{\eta}^w)$ per undistorted volume can be expanded into the Taylor series

$$F = \frac{1}{2} C_{ijkl}^{ab} \eta_{ij}^a \eta_{kl}^b + \frac{1}{6} C_{ijklmn}^{abc} \eta_{ij}^a \eta_{kl}^b \eta_{mn}^c + \dots = \frac{1}{2} C_{ij}^{ab} \eta_i^a \eta_j^b + \frac{1}{6} C_{ijk}^{abc} \eta_i^a \eta_j^b \eta_k^c + \dots \quad (3)$$

with C_{ijkl}^{ab} being second-order and C_{ijklmn}^{abc} third-order Cartesian elastic constants due to Brugger [16], which is perhaps the most familiar notation ($i, j, k, l, m, n \in \{1, 2, 3\}$; $a, b \in \{u, w\}$). In the right part of (3), the irreducible strain components of appendix A are used. Here we have, e.g., $i \in \{1, \dots, 6\}$ if $a = u$ and $i \in \{1, \dots, 9\}$ if $a = w$. Because of the index permutation symmetries and the symmetries of the QC, not all of these C are independent. If the elastic energy is to be written with independent elastic constants C_i^{ab} and C_i^{abc} of second and third order only, one has to use the invariants I_i^{ab} and I_i^{abc} of the generalized elasticity:

$$F = C_i^{ab} I_i^{ab} + C_i^{abc} I_i^{abc} + \dots \quad (4)$$

These invariants (and also expansions (3)) must fulfil the condition $I(g\boldsymbol{\eta}^u, g\boldsymbol{\eta}^w) = I(\boldsymbol{\eta}^u, \boldsymbol{\eta}^w)$ for any transformation g of the icosahedral group Y (or Y_h). Clearly, symmetries such as

$I_i^{uuw} = I_i^{uwu} = \dots$ and $C_i^{uuw} = C_i^{uwu} = \dots$ are assumed to hold. The elastic constants of (3) follow from appropriate repeated differentiation with respect to components of η^u and η^w .

A generalization of the classical result [16] shows that the generalized Piola–Kirchhoff stresses t^u and t^w , which are measured in the undistorted state, have components

$$t_{ij}^a = \frac{\partial F}{\partial \eta_{ij}^a} = C_{ijkl}^{ab} \eta_{kl}^b + \frac{1}{2} C_{ijklmn}^{abc} \eta_{kl}^b \eta_{mn}^c + \dots = C_k^{ab} \frac{\partial I_k^{ab}}{\partial \eta_{ij}^a} + C_k^{abc} \frac{\partial I_k^{abc}}{\partial \eta_{ij}^a} + \dots \quad (5)$$

The irreducible components of t^u and t^w have the same form as those of η^u and η^w , and it is $t_i^a = \frac{\partial F}{\partial \eta_i^a}$, where the possible pairs of (a, i) are again evident from appendix A. Cauchy stresses σ^u and σ^w are measured in the distorted state. They follow immediately from the Piola–Kirchhoff stresses [8]. Because E^\perp remains unchanged even for a finite deformation, the phasonic case must be treated with some care:

$$\sigma_{ij}^u = \Delta^{-1} F_{ik} F_{jl} t_{kl}^u \quad \sigma_{ij}^w = \Delta^{-1} F_{jk} t_{ik}^w \quad (6)$$

where $\Delta = \det F$.

Since our intention is to produce certain strains by means of applied stresses, we should rather work with the isothermal Gibbs enthalpy $G(t^u, t^w)$ [16],

$$G = S_i^{ab} I_i^{ab} + S_i^{abc} I_i^{abc} + \dots \quad (7)$$

S_i^{ab} and S_i^{abc} are independent elastic compliances, and the same invariants as in (4) appear, but this time formulated with components of t^u and t^w . One obtains the strain–stress relations

$$\eta_{ij}^a = -\frac{\partial G}{\partial t_{ij}^a} = -S_{ijkl}^{ab} t_{kl}^b - \frac{1}{2} S_{ijklmn}^{abc} t_{kl}^b t_{mn}^c + \dots = -S_k^{ab} \frac{\partial I_k^{ab}}{\partial t_{ij}^a} - S_k^{abc} \frac{\partial I_k^{abc}}{\partial t_{ij}^a} - \dots \quad (8)$$

The irreducible form hereof is obvious.

3. The elastic invariants

First thing is to note the transformation behaviour of the strain tensors [2], which follows from the transformation behaviour of vectors in E^\parallel and E^\perp :

$$u: (\Gamma^3 \otimes \Gamma^3)_s = \Gamma^1 \oplus \Gamma^5 \quad w: \Gamma^{3'} \otimes \Gamma^3 = \Gamma^4 \oplus \Gamma^5. \quad (9)$$

Index s means symmetrized. The irreducible components of the strain tensors are given in appendix A, and the associated transformation matrices are displayed in table A1. They are deduced from the transformation of the Cartesian strain tensors $\eta^a = \eta_{ij}^a \eta_{ij}^a$, where η_{ij}^a is a basis deformation with component ij being 1 and all others 0: $g\eta^a = \eta_{ij}^a g \eta_{ij}^a = (g\eta^a)_{ij} \eta_{ij}^a$, where $(g\eta^u)_{ij} = D_{ik}^3(g) D_{jl}^3(g) \eta_{kl}^u$, $(g\eta^w)_{ij} = D_{ik}^{3'}(g) D_{jl}^3(g) \eta_{kl}^w$ and $D^3(g)$, $D^{3'}(g)$ are the coordinate transformation matrices of table A1.

The characters of symmetrized product representations $D = (\tilde{D} \otimes \tilde{D})_s$ and $D = (\tilde{D} \otimes \tilde{D} \otimes \tilde{D})_s$, respectively, of one and the same representation \tilde{D} , which the terms occurring in (3) transform after, are

$$\begin{aligned} \chi^D(g) &= \frac{1}{2} [\chi^{\tilde{D}}(g)^2 + \chi^{\tilde{D}}(g^2)] \quad \text{and} \\ \chi^D(g) &= \frac{1}{6} [\chi^{\tilde{D}}(g)^3 + 3\chi^{\tilde{D}}(g^2)\chi^{\tilde{D}}(g) + 2\chi^{\tilde{D}}(g^3)]. \end{aligned} \quad (10)$$

Herewith, the following Clebsch–Gordan series for the representations acting in the respective symmetrized product spaces of second-order tensors are straightforward and given below in

Table 1. Tabulation of symmetrized third-order vector spaces. In the last column, all possible third-order expressions consisting of irreducible strains, referring to orthonormal basis sets, are listed.

Case	Dimension <i>dim</i> of symmetrized space	Components with respect to an orthonormal basis set
<i>uuu</i>	56	$(\eta_i^u)^3, \sqrt{3}(\eta_i^u)^2 \eta_j^u, \sqrt{6}\eta_i^u \eta_j^u \eta_k^u$ ($i \neq j \neq k \neq i$)
<i>uuw</i>	189	$(\eta_i^u)^2 \eta_k^w, \sqrt{2}\eta_i^u \eta_j^u \eta_k^w$ ($i \neq j$)
<i>uww</i>	270	$\eta_i^u (\eta_j^w)^2, \sqrt{2}\eta_i^u \eta_j^w \eta_k^w$ ($j \neq k$)
<i>www</i>	165	$(\eta_i^w)^3, \sqrt{3}(\eta_i^w)^2 \eta_j^w, \sqrt{6}\eta_i^w \eta_j^w \eta_k^w$ ($i \neq j \neq k \neq i$)

the order *uu, uw, ww, uuu, uuw, uww, www* [17]:

$$\begin{aligned}
 [(\Gamma^1 \oplus \Gamma^5) \otimes (\Gamma^1 \oplus \Gamma^5)]_s &= 2\Gamma^1 \oplus \Gamma^4 \oplus 3\Gamma^5 \\
 (\Gamma^1 \oplus \Gamma^5) \otimes (\Gamma^4 \oplus \Gamma^5) &= \Gamma^1 \oplus 2\Gamma^3 \oplus 2\Gamma^{3'} \oplus 4\Gamma^4 \oplus 5\Gamma^5 \\
 [(\Gamma^4 \oplus \Gamma^5) \otimes (\Gamma^4 \oplus \Gamma^5)]_s &= 2\Gamma^1 \oplus \Gamma^3 \oplus \Gamma^{3'} \oplus 3\Gamma^4 \oplus 5\Gamma^5 \\
 [(\Gamma^1 \oplus \Gamma^5) \otimes (\Gamma^1 \oplus \Gamma^5) \otimes (\Gamma^1 \oplus \Gamma^5)]_s &= 4\Gamma^1 \oplus \Gamma^3 \oplus \Gamma^{3'} \oplus 4\Gamma^4 \oplus 6\Gamma^5 \\
 [(\Gamma^1 \oplus \Gamma^5) \otimes (\Gamma^1 \oplus \Gamma^5)]_s \otimes (\Gamma^4 \oplus \Gamma^5) &= 4\Gamma^1 \oplus 8\Gamma^3 \oplus 8\Gamma^{3'} \oplus 13\Gamma^4 \oplus 17\Gamma^5 \\
 (\Gamma^1 \oplus \Gamma^5) \otimes [(\Gamma^4 \oplus \Gamma^5) \otimes (\Gamma^4 \oplus \Gamma^5)]_s &= 7\Gamma^1 \oplus 11\Gamma^3 \oplus 11\Gamma^{3'} \oplus 18\Gamma^4 \oplus 25\Gamma^5 \\
 [(\Gamma^4 \oplus \Gamma^5) \otimes (\Gamma^4 \oplus \Gamma^5) \otimes (\Gamma^4 \oplus \Gamma^5)]_s &= 5\Gamma^1 \oplus 7\Gamma^3 \oplus 7\Gamma^{3'} \oplus 12\Gamma^4 \oplus 14\Gamma^5.
 \end{aligned} \tag{11}$$

We see that we have the following numbers of invariants: 2 (*uu*), 1 (*uw*), 2 (*ww*), 4 (*uuu*), 4 (*uuw*), 7 (*uww*), 5 (*www*). These numbers have already been calculated in an earlier work [18].

The second-order invariants can readily be written as simple scalar products $\eta^{u,1} \cdot \eta^{u,1}, \eta^{u,5} \cdot \eta^{u,5}, \eta^{u,5} \cdot \eta^{w,5}, \eta^{w,4} \cdot \eta^{w,4}$ and $\eta^{w,5} \cdot \eta^{w,5}$ of vectors containing the irreducible strain components (see appendix A and [19]). Of course, most of the third-order invariants are more complicated.

In table 1, the symmetrized third-order vector spaces are specified in more detail. Some terms must be weighted with factors to become components for an orthonormal basis set and to transform orthogonal among all others. The third-order elastic invariants are the components for the basis vectors of the identity representation.

Basis vectors for an irreducible group representation α are obtained by means of the projection operators $P_{lk}^\alpha = \frac{d_\alpha}{|G|} \sum_{g \in G} D_{lk}^{\alpha*}(g) D(g)$ [20], with $D(g)$ being the linear operators acting on the full (reducible) vector space (see table 1). These projectors have the property $P_{lk}^\alpha e_{ij}^\beta = \delta_{\alpha\beta} \delta_{jk} e_{il}^\beta$. Here, e_{ij}^β is a basis vector transforming as the index j of the irreducible representation β , and $1 \leq i \leq n_D^\beta$, where n_D^β is the multiplicity of β in D . To split the full vector space into subspaces spanned by orthogonal irreducible basis vectors, the vectors $e_{i1}^\alpha \in \text{Im } P_{11}^\alpha, e_{i1}^\alpha \perp e_{j1}^\alpha$ for $i \neq j$, must be calculated. The other basis vectors are then $e_{ij}^\alpha = P_{j1}^\alpha e_{i1}^\alpha$, where $i \in \{1, \dots, n_D^\alpha\}, j \in \{2, \dots, d_\alpha\}$. The operators $D(g)$ are calculated as orthogonal transformation matrices for the components of table 1.

Herewith, the third-order invariants can be found directly from the full spaces of table 1. However, we picked the third-order invariants by the following procedure: calculate the second-order irreducible components for the irreducible representations Γ^1, Γ^4 and Γ^5 occurring in the first and third series of (11). Then set up all possible invariant scalar products

with irreducible components of η^u and η^w and throw away linearly occurring dependent invariants.

Since the components for different irreducible representations do not mix under transformation, another very elegant method, leading immediately to the ordering with respect to irreducible representations described below, is to expand the triple products of (11) completely, for example, $[(\Gamma^4 \oplus \Gamma^5) \otimes (\Gamma^4 \oplus \Gamma^5) \otimes (\Gamma^4 \oplus \Gamma^5)]_s = (\Gamma^4 \otimes \Gamma^4 \otimes \Gamma^4)_s \oplus [(\Gamma^4 \otimes \Gamma^4)_s \otimes \Gamma^5] \oplus [\Gamma^4 \otimes (\Gamma^5 \otimes \Gamma^5)]_s \oplus (\Gamma^5 \otimes \Gamma^5 \otimes \Gamma^5)_s$, and search the invariants for each of the arising triple products of irreducible representations separately.

The third-order invariants are listed in appendix B. They consist of components of as few different irreducible representations as possible (see table B1). For each of the four cases, they are orthonormal, i.e. $\sum_{k=1}^{dim} v_{i,k} v_{j,k} = \delta_{ij}$, where dim are the respective numbers in the second column of table 1 and $v_{i,k}$ is the coefficient of the third-order term k in the invariant i . Furthermore, we have tried, on the one hand, to choose the invariants as short as possible and, on the other hand, to bring them to a suitable form for comparing with each other and with the invariants of [14] (see appendices B and C for details).

4. Discussion

From (11), there are 20 independent third-order elastic invariants, or elastic constants, describing the non-linear elasticity of icosahedral QC. Since there exist four uuw -invariants, we have four non-linear phonon–phason couplings. The other third-order invariants are unsuitable if one wants to generate phason strains or stresses. Despite this, from all the third-order invariants, the four uuu ones will play the most important role because of their influence on the phonon wave propagation. To our knowledge, these third-order phonon elastic constants have not been determined for QC so far.

Generating phason Γ^4 -strains from phonon stresses now is possible through the invariant I_1^{uuw} . Due to (8),

$$\begin{aligned} \eta_1^w &= \frac{S_1^{uuw}}{20} \sqrt{30} \left[3 (t_2^u)^2 + 3 (t_3^u)^2 - 2 (t_4^u)^2 - 2 (t_5^u)^2 - 2 (t_6^u)^2 \right] \\ \eta_2^w &= -\frac{S_1^{uuw}}{2} \sqrt{3} \left(2t_3^u t_4^u + \sqrt{2} t_5^u t_6^u \right) \\ \eta_3^w &= \frac{S_1^{uuw}}{2} \left(3t_2^u t_5^u + \sqrt{3} t_3^u t_5^u - \sqrt{6} t_4^u t_6^u \right) \\ \eta_4^w &= -\frac{S_1^{uuw}}{2} \left(3t_2^u t_6^u - \sqrt{3} t_3^u t_6^u + \sqrt{6} t_4^u t_5^u \right). \end{aligned} \tag{12}$$

Here, a perhaps unexpected factor of 3 is present because of the summation rule in (8). From (12), it is obvious that phason Γ^4 -strains arise from shear stresses t_2^u, \dots, t_6^u . η_1^w is obtained, for example, by applying the stress $t_3^u \equiv t$ and all other $t_j^u = 0$, η_2^w by applying $t_3^u \equiv t, t_4^u \equiv \sqrt{3}/2t$, η_3^w by applying $t_3^u \equiv t, t_5^u \equiv \sqrt{3}/2t$ and η_4^w via $t_3^u \equiv t, t_6^u \equiv \sqrt{3}/2t$. The magnitude of an eventually existing phonon Γ^1 -stress, which is hydrostatic pressure t_1^u , has only an indirect effect on the phason Γ^4 strains, due to the pressure dependence of elastic compliances. Note that the Γ^4 -symmetry is unlikely to exist without simultaneous phason Γ^5 -symmetry, which is generated by shear stresses even in the linear, but also in the non-linear regime according to higher order compliances (see table B1).

Another possibility of obtaining phason Γ^4 -strains is, for example, the quartic electrostriction [22].

Table A1. Tabulation of the transformation matrices for the icosahedral irreducible representations. They are given for two appropriate generating elements of Y , i.e. a fivefold rotation C_5 and a threefold C_3 . $\tau - 1 = \frac{1}{\tau}$. For Y_h , the inversion i must be included. i does not change the strains [22].

Γ	$g = C_5$	$g = C_3$
$\Gamma^1 \equiv 1$	1	1
$\Gamma^3 \equiv 3$	$\frac{1}{2} \begin{pmatrix} \tau & \tau - 1 & -1 \\ \tau - 1 & 1 & \tau \\ 1 & -\tau & \tau - 1 \end{pmatrix}$	$\begin{pmatrix} 0 & 0 & 1 \\ 1 & 0 & 0 \\ 0 & 1 & 0 \end{pmatrix}$
$\Gamma^{3'} \equiv 3'$	$\frac{1}{2} \begin{pmatrix} 1 - \tau & -\tau & -1 \\ -\tau & 1 & 1 - \tau \\ 1 & \tau - 1 & -\tau \end{pmatrix}$	$\begin{pmatrix} 0 & 0 & 1 \\ 1 & 0 & 0 \\ 0 & 1 & 0 \end{pmatrix}$
$\Gamma^4 \equiv 4$	$\frac{1}{4} \begin{pmatrix} -1 & -\sqrt{5} & \sqrt{5} & -\sqrt{5} \\ -\sqrt{5} & -1 & -3 & -1 \\ -\sqrt{5} & 3 & 1 & -1 \\ \sqrt{5} & 1 & -1 & -3 \end{pmatrix}$	$\begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 1 & 0 \end{pmatrix}$
$\Gamma^5 \equiv 5$	$\frac{1}{4} \begin{pmatrix} 1 & -\sqrt{3} & -\sqrt{6} & 0 & \sqrt{6} \\ -\sqrt{3} & -1 & -\sqrt{2} & -2\sqrt{2} & -\sqrt{2} \\ -\sqrt{6} & -\sqrt{2} & 2 & 0 & 2 \\ 0 & 2\sqrt{2} & 0 & -2 & 2 \\ -\sqrt{6} & \sqrt{2} & -2 & 2 & 0 \end{pmatrix}$	$\frac{1}{2} \begin{pmatrix} -1 & -\sqrt{3} & 0 & 0 & 0 \\ \sqrt{3} & -1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 2 \\ 0 & 0 & 2 & 0 & 0 \\ 0 & 0 & 0 & 2 & 0 \end{pmatrix}$

Appendix A. Icosahedral irreducible strains

The icosahedral irreducible strain components given below are from [21]. They refer to the same coordinate systems as in [15, 19, 22]. In table A1, the icosahedral irreducible transformation matrices are given, which the irreducible strains and stresses transform after. Other coordinate systems in use are compared to ours in some detail in [15].

$$\begin{aligned}
 \eta^{u,1} &= \eta_1^u = \frac{1}{\sqrt{3}} (\eta_{11}^u + \eta_{22}^u + \eta_{33}^u) \\
 \eta^{u,5} &= \begin{pmatrix} \eta_2^u \\ \eta_3^u \\ \eta_4^u \\ \eta_5^u \\ \eta_6^u \end{pmatrix} = \begin{pmatrix} \frac{1}{2\sqrt{3}} (-\tau^2 \eta_{11}^u + \frac{1}{\tau^2} \eta_{22}^u + (\tau + \frac{1}{\tau}) \eta_{33}^u) \\ \frac{1}{2} (\frac{1}{\tau} \eta_{11}^u - \tau \eta_{22}^u + \eta_{33}^u) \\ \frac{1}{\sqrt{2}} (\eta_{12}^u + \eta_{21}^u) \\ \frac{1}{\sqrt{2}} (\eta_{23}^u + \eta_{32}^u) \\ \frac{1}{\sqrt{2}} (\eta_{31}^u + \eta_{13}^u) \end{pmatrix} \\
 \eta^{w,4} &= \begin{pmatrix} \eta_1^w \\ \eta_2^w \\ \eta_3^w \\ \eta_4^w \end{pmatrix} = \frac{1}{\sqrt{3}} \begin{pmatrix} \eta_{11}^w + \eta_{22}^w + \eta_{33}^w \\ \frac{1}{\tau} \eta_{21}^w + \tau \eta_{12}^w \\ \frac{1}{\tau} \eta_{32}^w + \tau \eta_{23}^w \\ \frac{1}{\tau} \eta_{13}^w + \tau \eta_{31}^w \end{pmatrix} \\
 \eta^{w,5} &= \begin{pmatrix} \eta_5^w \\ \eta_6^w \\ \eta_7^w \\ \eta_8^w \\ \eta_9^w \end{pmatrix} = \frac{1}{\sqrt{6}} \begin{pmatrix} \sqrt{3} (\eta_{11}^w - \eta_{22}^w) \\ \eta_{11}^w + \eta_{22}^w - 2\eta_{33}^w \\ \sqrt{2} (\tau \eta_{21}^w - \frac{1}{\tau} \eta_{12}^w) \\ \sqrt{2} (\tau \eta_{32}^w - \frac{1}{\tau} \eta_{23}^w) \\ \sqrt{2} (\tau \eta_{13}^w - \frac{1}{\tau} \eta_{31}^w) \end{pmatrix}.
 \end{aligned} \tag{A.1}$$

Appendix B. Third-order icosahedral invariants

$$\begin{aligned}
 I_1^{uuu} &= (\eta_1^u)^3 \\
 I_2^{uuu} &= \frac{1}{20}\sqrt{30}\left[-(\eta_3^u)^3 + \eta_3^u (\eta_5^u)^2 + \eta_3^u (\eta_6^u)^2 - 2\eta_3^u (\eta_4^u)^2 + 3\eta_3^u (\eta_2^u)^2\right. \\
 &\quad \left.+ 4\sqrt{2}\eta_4^u \eta_5^u \eta_6^u + \sqrt{3}\eta_2^u (\eta_5^u)^2 - \sqrt{3}\eta_2^u (\eta_6^u)^2\right] \\
 I_3^{uuu} &= \frac{1}{20}\sqrt{10}\left[(\eta_2^u)^3 - 3\eta_2^u (\eta_3^u)^2 - 3\eta_2^u (\eta_5^u)^2 - 3\eta_2^u (\eta_6^u)^2 + 6\eta_2^u (\eta_4^u)^2\right. \\
 &\quad \left.+ 3\sqrt{3}\eta_3^u (\eta_5^u)^2 - 3\sqrt{3}\eta_3^u (\eta_6^u)^2\right] \\
 I_4^{uuu} &= \frac{1}{5}\sqrt{15}\eta_1^u \left[(\eta_2^u)^2 + (\eta_3^u)^2 + (\eta_4^u)^2 + (\eta_5^u)^2 + (\eta_6^u)^2\right]
 \end{aligned} \tag{B.1}$$

$$\begin{aligned}
 I_1^{uww} &= \frac{1}{60}\sqrt{30}\left[2(\eta_4^u)^2 \eta_1^w + 2(\eta_5^u)^2 \eta_1^w + 2(\eta_6^u)^2 \eta_1^w - 3(\eta_2^u)^2 \eta_1^w - 3(\eta_3^u)^2 \eta_1^w\right. \\
 &\quad \left.+ 2\sqrt{5}\eta_5^u \eta_6^u \eta_2^w + 2\sqrt{5}\eta_4^u \eta_6^u \eta_3^w + 2\sqrt{5}\eta_4^u \eta_5^u \eta_4^w - \sqrt{10}\eta_3^u \eta_5^u \eta_3^w\right. \\
 &\quad \left.- \sqrt{10}\eta_3^u \eta_6^u \eta_4^w + 2\sqrt{10}\eta_3^u \eta_4^u \eta_2^w - \sqrt{30}\eta_2^u \eta_5^u \eta_3^w + \sqrt{30}\eta_2^u \eta_6^u \eta_4^w\right] \\
 I_2^{uww} &= \frac{1}{60}\sqrt{30}\left[(\eta_5^u)^2 \eta_6^w + (\eta_6^u)^2 \eta_6^w - 2(\eta_4^u)^2 \eta_6^w + 2\eta_3^u \eta_5^u \eta_8^w + 2\eta_3^u \eta_6^u \eta_9^w + 3(\eta_2^u)^2 \eta_6^w\right. \\
 &\quad \left.- 3(\eta_3^u)^2 \eta_6^w - 4\eta_3^u \eta_4^u \eta_7^w + 6\eta_2^u \eta_3^u \eta_5^w + 4\sqrt{2}\eta_5^u \eta_6^u \eta_7^w + 4\sqrt{2}\eta_4^u \eta_6^u \eta_8^w\right. \\
 &\quad \left.+ 4\sqrt{2}\eta_4^u \eta_5^u \eta_9^w + \sqrt{3}(\eta_5^u)^2 \eta_5^w - \sqrt{3}(\eta_6^u)^2 \eta_5^w + 2\sqrt{3}\eta_2^u \eta_5^u \eta_8^w - 2\sqrt{3}\eta_2^u \eta_6^u \eta_9^w\right] \\
 I_3^{uww} &= \frac{1}{20}\sqrt{10}\left[(\eta_2^u)^2 \eta_5^w - (\eta_3^u)^2 \eta_5^w - (\eta_5^u)^2 \eta_5^w - (\eta_6^u)^2 \eta_5^w + 2(\eta_4^u)^2 \eta_5^w - 2\eta_2^u \eta_3^u \eta_6^w\right. \\
 &\quad \left.- 2\eta_2^u \eta_5^u \eta_8^w - 2\eta_2^u \eta_6^u \eta_9^w + 4\eta_2^u \eta_4^u \eta_7^w + \sqrt{3}(\eta_5^u)^2 \eta_6^w - \sqrt{3}(\eta_6^u)^2 \eta_6^w\right. \\
 &\quad \left.+ 2\sqrt{3}\eta_3^u \eta_5^u \eta_8^w - 2\sqrt{3}\eta_3^u \eta_6^u \eta_9^w\right] \\
 I_4^{uww} &= \frac{1}{5}\sqrt{10}\eta_1^u (\eta_2^u \eta_5^w + \eta_3^u \eta_6^w + \eta_4^u \eta_7^w + \eta_5^u \eta_8^w + \eta_6^u \eta_9^w)
 \end{aligned} \tag{B.2}$$

$$\begin{aligned}
 I_1^{uww} &= \frac{1}{2}\eta_1^u \left[(\eta_1^w)^2 + (\eta_2^w)^2 + (\eta_3^w)^2 + (\eta_4^w)^2\right] \\
 I_2^{uww} &= \frac{1}{5}\sqrt{5}\eta_1^u \left[(\eta_5^w)^2 + (\eta_6^w)^2 + (\eta_7^w)^2 + (\eta_8^w)^2 + (\eta_9^w)^2\right] \\
 I_3^{uww} &= \frac{1}{60}\sqrt{30}\left[\eta_3^u (\eta_8^w)^2 + \eta_3^u (\eta_9^w)^2 - 2\eta_3^u (\eta_7^w)^2 + 2\eta_5^u \eta_6^w \eta_8^w + 2\eta_6^u \eta_6^w \eta_9^w + 3\eta_3^u (\eta_5^w)^2\right. \\
 &\quad \left.- 3\eta_3^u (\eta_6^w)^2 - 4\eta_4^u \eta_6^w \eta_7^w + 6\eta_2^u \eta_5^w \eta_6^w + 4\sqrt{2}\eta_5^u \eta_7^w \eta_9^w + 4\sqrt{2}\eta_6^u \eta_7^w \eta_8^w\right. \\
 &\quad \left.+ 4\sqrt{2}\eta_4^u \eta_8^w \eta_9^w - \sqrt{3}\eta_2^u (\eta_9^w)^2 + \sqrt{3}\eta_2^u (\eta_8^w)^2 - 2\sqrt{3}\eta_6^u \eta_5^w \eta_9^w + 2\sqrt{3}\eta_5^u \eta_5^w \eta_8^w\right] \\
 I_4^{uww} &= \frac{1}{20}\sqrt{10}\left[\eta_2^u (\eta_5^w)^2 - \eta_2^u (\eta_6^w)^2 - \eta_2^u (\eta_8^w)^2 - \eta_2^u (\eta_9^w)^2 + 2\eta_2^u (\eta_7^w)^2 - 2\eta_3^u \eta_5^w \eta_6^w\right. \\
 &\quad \left.- 2\eta_5^u \eta_5^w \eta_8^w - 2\eta_6^u \eta_5^w \eta_9^w + 4\eta_4^u \eta_5^w \eta_7^w + \sqrt{3}\eta_3^u (\eta_8^w)^2 - \sqrt{3}\eta_3^u (\eta_9^w)^2\right. \\
 &\quad \left.+ 2\sqrt{3}\eta_5^u \eta_6^w \eta_8^w - 2\sqrt{3}\eta_6^u \eta_6^w \eta_9^w\right] \\
 I_5^{uww} &= \frac{1}{30}\sqrt{15}\left[2\eta_6^u \eta_2^w \eta_3^w + 2\eta_5^u \eta_2^w \eta_4^w + 2\eta_4^u \eta_3^w \eta_4^w + \sqrt{2}\eta_3^u (\eta_3^w)^2 + \sqrt{2}\eta_3^u (\eta_4^w)^2\right. \\
 &\quad \left.- 2\sqrt{2}\eta_3^u (\eta_2^w)^2 + 2\sqrt{5}\eta_4^u \eta_1^w \eta_2^w + 2\sqrt{5}\eta_5^u \eta_1^w \eta_3^w + 2\sqrt{5}\eta_6^u \eta_1^w \eta_4^w\right. \\
 &\quad \left.+ \sqrt{6}\eta_2^u (\eta_3^w)^2 - \sqrt{6}\eta_2^u (\eta_4^w)^2\right]
 \end{aligned} \tag{B.3}$$

$$\begin{aligned}
I_6^{uww} &= \frac{1}{60}\sqrt{15} \left(4\eta_4^u \eta_1^w \eta_7^w + 4\eta_5^u \eta_1^w \eta_8^w + 4\eta_6^u \eta_1^w \eta_9^w - 6\eta_2^u \eta_1^w \eta_5^w - 6\eta_3^u \eta_1^w \eta_6^w \right. \\
&\quad + 2\sqrt{5}\eta_6^u \eta_2^w \eta_8^w + 2\sqrt{5}\eta_5^u \eta_2^w \eta_9^w + 2\sqrt{5}\eta_6^u \eta_3^w \eta_7^w + 2\sqrt{5}\eta_4^u \eta_3^w \eta_9^w \\
&\quad + 2\sqrt{5}\eta_5^u \eta_4^w \eta_7^w + 2\sqrt{5}\eta_4^u \eta_4^w \eta_8^w - \sqrt{10}\eta_5^u \eta_3^w \eta_6^w - \sqrt{10}\eta_3^u \eta_3^w \eta_8^w \\
&\quad - \sqrt{10}\eta_6^u \eta_4^w \eta_6^w - \sqrt{10}\eta_3^u \eta_4^w \eta_9^w + 2\sqrt{10}\eta_4^u \eta_2^w \eta_6^w + 2\sqrt{10}\eta_3^u \eta_2^w \eta_7^w \\
&\quad \left. - \sqrt{30}\eta_5^u \eta_3^w \eta_5^w - \sqrt{30}\eta_2^u \eta_3^w \eta_8^w + \sqrt{30}\eta_6^u \eta_4^w \eta_5^w + \sqrt{30}\eta_2^u \eta_4^w \eta_9^w \right) \\
I_7^{uww} &= \frac{1}{20}\sqrt{10} \left(\eta_5^u \eta_3^w \eta_5^w - \eta_2^u \eta_3^w \eta_8^w + \eta_6^u \eta_4^w \eta_5^w - \eta_2^u \eta_4^w \eta_9^w - 2\eta_4^u \eta_2^w \eta_5^w + 2\eta_2^u \eta_2^w \eta_7^w \right. \\
&\quad - \sqrt{3}\eta_5^u \eta_3^w \eta_6^w + \sqrt{3}\eta_3^u \eta_3^w \eta_8^w + \sqrt{3}\eta_6^u \eta_4^w \eta_6^w - \sqrt{3}\eta_3^u \eta_4^w \eta_9^w \\
&\quad + \sqrt{6}\eta_6^u \eta_2^w \eta_8^w - \sqrt{6}\eta_5^u \eta_2^w \eta_9^w - \sqrt{6}\eta_6^u \eta_3^w \eta_7^w + \sqrt{6}\eta_4^u \eta_3^w \eta_9^w \\
&\quad \left. + \sqrt{6}\eta_3^u \eta_4^w \eta_7^w - \sqrt{6}\eta_4^u \eta_4^w \eta_8^w - \sqrt{10}\eta_3^u \eta_1^w \eta_5^w + \sqrt{10}\eta_2^u \eta_1^w \eta_6^w \right) \\
I_1^{www} &= \frac{1}{4}\sqrt{3} \left[(\eta_1^w)^3 - \eta_1^w (\eta_2^w)^2 - \eta_1^w (\eta_3^w)^2 - \eta_1^w (\eta_4^w)^2 + 2\sqrt{5}\eta_2^w \eta_3^w \eta_4^w \right] \\
I_2^{www} &= \frac{1}{20}\sqrt{30} \left[-(\eta_6^w)^3 + \eta_6^w (\eta_8^w)^2 + \eta_6^w (\eta_9^w)^2 - 2\eta_6^w (\eta_7^w)^2 + 3\eta_6^w (\eta_5^w)^2 \right. \\
&\quad \left. + 4\sqrt{2}\eta_7^w \eta_8^w \eta_9^w + \sqrt{3}\eta_5^w (\eta_8^w)^2 - \sqrt{3}\eta_5^w (\eta_9^w)^2 \right] \\
I_3^{www} &= \frac{1}{20}\sqrt{10} \left[(\eta_5^w)^3 - 3\eta_5^w (\eta_6^w)^2 - 3\eta_5^w (\eta_8^w)^2 - 3\eta_5^w (\eta_9^w)^2 + 6\eta_5^w (\eta_7^w)^2 \right. \\
&\quad \left. + 3\sqrt{3}\eta_6^w (\eta_8^w)^2 - 3\sqrt{3}\eta_6^w (\eta_9^w)^2 \right] \\
I_4^{www} &= \frac{1}{10}\sqrt{10} \left[(\eta_3^w)^2 \eta_6^w + (\eta_4^w)^2 \eta_6^w - 2(\eta_2^w)^2 \eta_6^w + \sqrt{2}\eta_2^w \eta_3^w \eta_9^w + \sqrt{2}\eta_2^w \eta_4^w \eta_8^w \right. \\
&\quad + \sqrt{2}\eta_3^w \eta_4^w \eta_7^w + \sqrt{3}(\eta_3^w)^2 \eta_5^w - \sqrt{3}(\eta_4^w)^2 \eta_5^w + \sqrt{10}\eta_1^w \eta_4^w \eta_9^w \\
&\quad \left. + \sqrt{10}\eta_1^w \eta_2^w \eta_7^w + \sqrt{10}\eta_1^w \eta_3^w \eta_8^w \right] \tag{B.4} \\
I_5^{www} &= \frac{1}{20}\sqrt{10} \left[2\eta_1^w (\eta_7^w)^2 + 2\eta_1^w (\eta_8^w)^2 + 2\eta_1^w (\eta_9^w)^2 - 3\eta_1^w (\eta_5^w)^2 - 3\eta_1^w (\eta_6^w)^2 \right. \\
&\quad + 2\sqrt{5}\eta_2^w \eta_8^w \eta_9^w + 2\sqrt{5}\eta_3^w \eta_7^w \eta_9^w + 2\sqrt{5}\eta_4^w \eta_7^w \eta_8^w - \sqrt{10}\eta_3^w \eta_6^w \eta_8^w \\
&\quad \left. - \sqrt{10}\eta_4^w \eta_6^w \eta_9^w + 2\sqrt{10}\eta_2^w \eta_6^w \eta_7^w - \sqrt{30}\eta_3^w \eta_5^w \eta_8^w + \sqrt{30}\eta_4^w \eta_5^w \eta_9^w \right].
\end{aligned}$$

The invariants comprise strain components of irreducible representations as displayed in table B1. Note that the following pairs of invariants have exactly the same structure: I_2^{uuu}/I_2^{www} , I_3^{uuu}/I_3^{www} , I_4^{uuu}/I_2^{www} , I_1^{uuw}/I_5^{www} , I_2^{uuw}/I_3^{www} , I_3^{uuw}/I_4^{www} , I_5^{uuw}/I_4^{www} . Furthermore, $3I_3^{uuw}$ is obtained from I_2^{www} by replacing all products $\eta_i^w \eta_j^w \eta_k^w$ by $\eta_{i-3}^u \eta_j^u \eta_k^u + \eta_i^w \eta_{j-3}^w + \eta_i^w \eta_j^w \eta_{k-3}^w$. The same is true for $3I_4^{uuw}$ and I_3^{www} . $\sqrt{6}I_6^{uuw}$ follows from I_5^{www} by replacing only the phason Γ^5 -components in the manner described above.

Appendix C. Relationship to other third-order invariants and elastic isotropy in phonon space

$$\begin{aligned}
\text{tr}(\eta^u)^3 &= 3\sqrt{3}I_1^{uuu} \\
\text{tr}(\eta^u) \{ \text{tr}[(\eta^u)^2] \}_s &= \sqrt{3}I_1^{uuu} + \sqrt{5}I_4^{uuu} \\
\{ \text{tr}[(\eta^u)^3] \}_s &= \frac{1}{3}\sqrt{3}I_1^{uuu} + \frac{1}{4}\sqrt{30}I_2^{uuu} - \frac{5}{12}\sqrt{6}I_3^{uuu} + \sqrt{5}I_4^{uuu} \tag{C.1}
\end{aligned}$$

Table B1. Powers of irreducible representation components $\eta_i^{a,\alpha}$ in third-order invariants. Possible combinations are: $\alpha = 1, i = 1$; $\alpha = 5, i = 2, 3, 4, 5, 6$ ($a = u$); $\alpha = 4, i = 1, 2, 3, 4$; $\alpha = 5, i = 5, 6, 7, 8, 9$ ($a = w$).

(a, α)	$(u, 1)$	$(u, 5)$	$(w, 4)$	$(w, 5)$	$(u, 1)$	$(u, 5)$	$(w, 4)$	$(w, 5)$
I_1^{uuu}	3				I_3^{uww}	1		2
I_2^{uuu}		3			I_4^{uww}	1		2
I_3^{uuu}		3			I_5^{uww}	1	2	
I_4^{uuu}	1	2			I_6^{uww}	1	1	1
I_1^{uuw}		2	1		I_7^{uww}	1	1	1
I_2^{uww}		2		1	I_1^{www}		3	
I_3^{uww}		2		1	I_2^{www}			3
I_4^{uww}	1	1		1	I_3^{www}			3
I_1^{uww}	1		2		I_4^{www}		2	1
I_2^{uww}	1			2	I_5^{www}		1	2

$$[\det(\boldsymbol{\eta}^u)]_s = \frac{1}{9}\sqrt{3}I_1^{uuu} + \frac{1}{12}\sqrt{30}I_2^{uuu} - \frac{5}{36}\sqrt{6}I_3^{uuu} - \frac{1}{6}\sqrt{5}I_4^{uuu}$$

$$\det(\boldsymbol{\eta}^w) = \frac{4}{9}I_1^{www} + \frac{2}{9}\sqrt{5}I_2^{www} - \frac{1}{9}\sqrt{15}I_4^{www} + \frac{1}{9}\sqrt{30}I_5^{www}.$$

In (C.1), index s denotes that η_{ij}^u must be replaced by $\frac{1}{2}(\eta_{ij}^u + \eta_{ji}^u)$. Some of our third-order invariants are just proportional to η_1^u times one of the second-order, invariant scalar products given in section 3. Note that $\text{tr}(\boldsymbol{\eta}^w)$ is not invariant. Our invariants I_1^{uuu} and I_4^{uuu} are also O(3)-invariants, while I_2^{uuu} and I_3^{uuu} must be combined to $\frac{3}{14}\sqrt{14}I_2^{uuu} - \frac{1}{14}\sqrt{70}I_3^{uuu}$ to give a linearly independent third O(3)-invariant. In the degenerate case of phononic isotropy, our elastic constants C_i^{uuu} can be expressed by sets of classical third-order elastic constants already in use: $C_1^{uuu} = \sqrt{3}(l + \frac{1}{9}n)$, $C_2^{uuu} = \frac{1}{12}\sqrt{30}n$, $C_3^{uuu} = -\frac{5}{36}\sqrt{6}n$, $C_4^{uuu} = \sqrt{5}(m - \frac{1}{6}n)$ [9, 13], $C_1^{uww} = \sqrt{3}(\frac{1}{2}\nu_1 + \nu_2 + \frac{4}{9}\nu_3)$, $C_2^{uww} = \frac{1}{3}\sqrt{30}\nu_3$, $C_3^{uww} = -\frac{5}{9}\sqrt{6}\nu_3$, $C_4^{uww} = \sqrt{5}(\nu_2 + \frac{4}{3}\nu_3)$ [23].

Up to normalization factors, the phason third-order invariants $\mathcal{I}'_5, \mathcal{I}_5, \mathcal{J}$ and \mathcal{J}' of Ishii [14] are transformed into our $I_2^{www}, I_3^{www}, I_4^{www}$ and I_5^{www} , respectively, by the substitutions $\eta_5^w \rightarrow -\frac{1}{4}(\sqrt{10}\eta_5^w + \sqrt{6}\eta_6^w)$ and $\eta_6^w \rightarrow \frac{1}{4}(-\sqrt{6}\eta_5^w + \sqrt{10}\eta_6^w)$. This is necessary because in [14] other irreducible components are used than in [21].

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