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Isotropic and anisotropic physical properties of quasicrystals

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Abstract. Since quasicrystals have positional and orientational long-range order, they are essentially anisotropic. However, the researches show that some physical properties of quasicrystals are isotropic. On the other hand, quasicrystals have additional phason degrees of freedom which can influence on their physical behaviours. To reveal the quasicrystal anisotropy, we investigate the quasicrystal elasticity and other physical properties, such as thermal expansion, piezoelectric and piezoresistance, for which one must consider the contributions of the phason field. The results indicate that: for the elastic properties, within linear phonon domain all quasicrystals are isotropic, and within nonlinear phonon domain the planar quasicrystals are still isotropic but the icosahedral quasicrystals are anisotropic. Moreover, the nonlinear elastic properties due to the coupling between phonons and phasons may reveal the anisotropic structure of QCs. For the other physical properties all quasicrystals behave like isotropic media except for piezoresistance properties of icosahedral quasicrystals due to the phason field.

PACS. 61.44.Br Quasicrystals – 62.20.Dc Elasticity, elastic constants

1 Introduction

The quasicrystals (QCs), possessing the aperiodic long-range order and noncrystallographic rotational symmetry, are considered as an individual class of material different from the two types of traditional solid, crystals and amorphous. QCs are fundamentally anisotropic because of their positional and orientational long-range order. Thus, the physical properties are expected to be anisotropic. Owing to important role playing in structural application, elastic properties of QCs are continually concerned. Several researches show that within linear phonon elasticity QCs behave essentially like isotropic media [1–5], but recent studies of *i*-Al-Pd-Mn demonstrated that the anisotropy can be detected by the nonlinear elastic properties due to pure phonon strain [6–8]. This can be explained by Hermann's Theorem. Hermann's Theorem shows that if an r-rank tensor has an N-fold symmetry axis and $r < N$, then this tensor also has a symmetry axis of an infinite order. Namely, a tensor is not anisotropic unless its rank $r \geqslant N$. As we know, the linear phonon elasticity is a fourth-rank tensor (so called, second-order elastic constant) and the fold of symmetry axes which QCs have are generally so high (> 4) that it must lead to the ∞-fold symmetry axes of the elastic constants [9–11]. In the view of this point, it is necessary to investigate nonlinear elasticity for the anisotropic structure of quasicrystals.

According to Landau's theory, the mass density is

$$\rho(\mathbf{r}) = \sum \rho_{\mathbf{G}} \mathbf{e}^{\mathbf{i}(\mathbf{G}\cdot\mathbf{r})+\phi_{\mathbf{G}}} \tag{1}$$

for quasicrystals, where $\mathbf{G}$ is reciprocal vector and ϕ_G is its phase. The phase can be expressed as the function of two displacements, the phonon field $\mathbf{u}$ and the phason field $\mathbf{w}$ [12],

$$\phi_G = \mathbf{G}^{\parallel} \cdot \mathbf{u} + \mathbf{G}^{\perp} \cdot \mathbf{w} \tag{2}$$

where $\mathbf{G}^{\parallel}$ and $\mathbf{u}$ are the reciprocal and phonon displacement vectors in the physical space, and $\mathbf{G}^{\perp}$ and $\mathbf{w}$ are the conjugate and phason displacement vectors in the perpendicular space. Therefore, in addition to the phonon, there exists another special elementary excitation (phason) in the quasicrystalline structure. In fact the phason degree of freedom is frozen at low temperature, but it can be activated and influence strongly the physical properties of QCs at high temperature. In this case, the total contribution to the QCs anisotropy should include both phonon and phason excitations. Several researchers have proved this point [11,13,14]. Thus, at this time Hermann's Theorem is valid for the phason degree of freedom as well.

In addition to elasticity, the situation also occurs to some other physical property tensors for which one must consider the contributions not only of the phonon field but also of the phason field. There are generally three types which are thermal expansion, piezoelectric and piezoresistance constants. So far, many experimental data on thermal expansion of quasicrystals have been reported, for instant, icosahedral QCs by Kupsch et al. [15], Swenson et al. [16], and decagonal QCs by Kupsch et al. [17], Inaba et al. [18]. Besides, theoretical studies on thermodynamics of equilibrium and transport properties of quasicrystals [19–21] have been carried out.

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The aim of this paper is to discuss the anisotropy of the physical properties of QCs. Section 2 focuses on the anisotropy of the quasicrystal elasticity. Section 3 goes into the anisotropies of thermal expansion, piezoelectric and piezoresistance constants. Section 4 draws some conclusions.

2 Elastic properties of quasicrystals

For QCs there exist two kinds of hydrodynamic variables: the phonon variable $\mathbf{u}$ and the phason variable $\mathbf{w}$, and then two kinds of strains: the phonon strain $E_{ij} = \frac{1}{2}(\partial_j u_i + \partial_i u_j)$ and the phason strain $W_{\alpha i} = \partial_i w_\alpha$, where subscripts $i, j, k, ...$ are used for coordinate components in physical space and subscripts $\alpha, \beta, \gamma, ...$ for coordinate components in the perpendicular space. It should be noted that the phonon variable transforms under the vector representation, whereas the phason variable transforms under another irreducible representation. The elastic energy density F which is a function of phonon and phason strains can be expanded into the Taylor series in the vicinity of $E_{ij} = 0$ and $W_{\alpha i} = 0$ to third order:

$$\begin{aligned} F = {} & \tfrac{1}{2}C_{ijkl}E_{ij}E_{kl} + \tfrac{1}{2}K_{\alpha i\beta j}W_{\alpha i}W_{\beta j} + R_{ij\alpha k}E_{ij}W_{\alpha k} \\ & + \tfrac{1}{6}C_{ijklmn}E_{ij}E_{kl}E_{mn} + \tfrac{1}{6}K_{\alpha i\beta j\gamma k}W_{\alpha i}W_{\beta j}W_{\gamma k} \\ & + \tfrac{1}{2}R^{(1)}_{ijkl\alpha m}E_{ij}E_{kl}W_{\alpha m} + \tfrac{1}{2}R^{(2)}_{ij\alpha k\beta l}E_{ij}W_{\alpha k}W_{\beta l} \end{aligned} \tag{3}$$

where

$$\begin{aligned} C_{ijkl} &= \left(\frac{\partial^2 F}{\partial E_{ij}\partial E_{kl}}\right)_0, \\ K_{\alpha i\beta j} &= \left(\frac{\partial^2 F}{\partial W_{\alpha i}\partial W_{\beta j}}\right)_0, \\ R_{ij\alpha k} &= \left(\frac{\partial^2 F}{\partial E_{ij}\partial W_{\alpha k}}\right)_0 \end{aligned} \tag{4}$$

are the second-order elastic constants of phonon field, phason field and phonon-phason coupling, respectively.

$$\begin{aligned} C_{ijklmn} &= \left(\frac{\partial^3 F}{\partial E_{ij}\partial E_{kl}\partial E_{mn}}\right)_0, \\ K_{\alpha i\beta j\gamma k} &= \left(\frac{\partial^3 F}{\partial W_{\alpha i}\partial W_{\beta j}\partial W_{\gamma k}}\right)_0, \\ R^{(1)}_{ijkl\alpha m} &= \left(\frac{\partial^3 F}{\partial E_{ij}\partial E_{kl}\partial W_{\alpha m}}\right)_0, \\ R^{(2)}_{ij\alpha k\beta l} &= \left(\frac{\partial^3 F}{\partial E_{ij}\partial W_{\alpha k}\partial W_{\beta l}}\right)_0 \end{aligned} \tag{5}$$

are the third-order elastic constants of phonon field, phason field, phonon-phonon-phason coupling, and phonon-phason-phason coupling, respectively.

Because the elastic property does not depend on the direction for 3D isotropic media, their symmetry group is SO(3). The dimension of the (phonon) strain tensor space is 6 for 3D isotropic media, when the deduced indexes:

$$\begin{aligned} (ij) &= 11, 22, 33, 23, 31, 12, \\ i &= 1,\ \ 2,\ \ 3,\ \ 4,\ \ 5,\ \ 6, \end{aligned} \tag{6}$$

are used. In the language of Young's diagrams [22], the strain tensor can symbolically denoted by a square $\square$. The linear and nonlinear elastic constant tensors can be graphically written as

$$\begin{aligned} &\{\underset{6}{\square} \otimes \underset{6}{\square}\}_s = \underset{21}{\square\square} \\ &\{\underset{6}{\square} \otimes \underset{6}{\square} \otimes \underset{6}{\square}\}_s = \underset{56}{\square\square\square}. \end{aligned} \tag{7}$$

The number below the equation is the dimension of the corresponding representation of SO(3). Using the formulae for calculating the characters of symmetric representations [20]:

$$\begin{aligned} \chi_{(A\times A)_s}(g) &= \frac{1}{2}[\chi_A(g)^2 + \chi_A(g^2)], \\ \chi_{(A\times A\times A)_s}(g) &= \frac{1}{6}[\chi_A(g)^3 + 3\chi_A(g)\chi_A(g^2) + 2\chi_A(g^3)], \end{aligned} \tag{8}$$

we can obtain by the formulae (7)

$$\begin{aligned} &\{(D_0 + D_2) \times (D_0 + D_2)\}_s = 2D_0 + 2D_2 + D_4, \\ &\{(D_0 + D_2) \times (D_0 + D_2) \times (D_0 + D_2)\}_s = \\ &\qquad 3D_0 + 3D_2 + D_3 + 2D_4 + D_6. \end{aligned} \tag{9}$$

where D_l is $(2l+1)$-dimensional irreducible representation of SO(3). Its representation space can be formed by usual spherical harmonics (Y_{lm}). Such reduction is often called Clebsh-Gordan series. From equation (9) it follows that there are two independent components of the linear elastic constant and three independent components of nonlinear elastic constant in 3D isotropic media.

Crystals are regarded as anisotropic media. The elastic properties of crystals have already been well-known. For example, cubic crystals have 3 independent components of linear elastic constant and 6 independent components of nonlinear elastic constant [10].

A three-dimensional QC is usually referred to icosahedral QCs. In the case of icosahedral QCs, $\mathbf{u}$ transforms under Γ_3 and $\mathbf{w}$ transforms under Γ_3' [23]. Then the phonon and phason strains transform under

$$(\Gamma_3 \times \Gamma_3)_s = \Gamma_1 + \Gamma_5, \Gamma_3 \times \Gamma_3' = \Gamma_4 + \Gamma_5. \tag{10}$$

The linear elastic tensor representations can be decomposed into

$$\begin{aligned} &\{(\Gamma_1 + \Gamma_5) \times (\Gamma_1 + \Gamma_5)\}_s = 2\Gamma_1 + \Gamma_4 + 3\Gamma_5, \\ &\{(\Gamma_4 + \Gamma_5) \times (\Gamma_4 + \Gamma_5)\}_s = 2\Gamma_1 + \Gamma_3 + \Gamma_3' + 3\Gamma_4 + 5\Gamma_5, \\ &\quad (\Gamma_1 + \Gamma_5) \times (\Gamma_4 + \Gamma_5) = \Gamma_1 + 2\Gamma_3 + 2\Gamma_3' + 4\Gamma_4 + 5\Gamma_5. \end{aligned} \tag{11}$$

This means that within linear elasticity icosahedral QCs have two independent elastic constants due to the phonon field (uu), two independent elastic constants due to the phason field (ww) and one independent constants associated with the pnonon-phason coupling (uw). From the

Table 1. The number of independent elastic constants for various solids. n_{uu} is the number of independent linear phonon elastic constants, n_{ww} is the number of independent linear phason elastic constants, n_{uw} is the number of independent linear elastic constants due to phonon-phason coupling, n_{uuu} is the number of independent nonlinear phonon elastic constants, n_{www} is the number of independent nonlinear phason elastic constants, n_{uuw} is the number of independent nonlinear elastic constants due to phonon-phonon-phason coupling, n_{uww} is the number of independent nonlinear elastic constants due to phonon-phason-phason coupling.

	n_{uu}	n_{uw}	n_{ww}	n_{uuu}	n_{uuw}	n_{uww}	n_{www}
3D isotropic media	2	–	–	3	–	–	–
Cubic crystals	3	–	–	6	–	–	–
Icosahedral QCs	2	1	2	4	4	7	5
2D isotropic media	2	–	–	2	–	–	–
Decagonal QCs	2	2	2	2	4	6	4
	2	1	2	2	2	4	2
Octagonal QCs	2	2	4	2	6	8	4
	2	1	3	2	3	5	2
Dodecagonal QCs	2	0	4	2	2	8	2
	2	0	3	2	1	5	1

reductions

$$\begin{aligned}&\{(\Gamma_1+\Gamma_5)\times(\Gamma_1+\Gamma_5)\times(\Gamma_1+\Gamma_5)\}_s=\\&\quad 4\Gamma_1+\Gamma_3+\Gamma_3'+4\Gamma_4+6\Gamma_5,\\&\{(\Gamma_4+\Gamma_5)\times(\Gamma_4+\Gamma_5)\times(\Gamma_4+\Gamma_5)\}_s=\\&\quad 5\Gamma_1+7\Gamma_3+7\Gamma_3'+12\Gamma_4+14\Gamma_5,\\&\{(\Gamma_1+\Gamma_5)\times(\Gamma_1+\Gamma_5)\}_s\times(\Gamma_4+\Gamma_5)=\\&\quad 4\Gamma_1+8\Gamma_3+8\Gamma_3'+13\Gamma_4+17\Gamma_5,\\&(\Gamma_1+\Gamma_5)\times\{(\Gamma_4+\Gamma_5)\times(\Gamma_4+\Gamma_5)\}_s=\\&\quad 7\Gamma_1+11\Gamma_3+11\Gamma_3'+18\Gamma_4+25\Gamma_5,\end{aligned}\qquad(12)$$

we can get that with nonlinear elasticity icosahedral QCs have 20 independent (third-order) elastic constants: four are due to the phonon field (uuu), five due to the phason field (www), four due to the phonon-phonon-phason coupling (uuw) and seven due to the phonon-phason-phason coupling (uww) [8]. Using the same method, we can deduce the number of independent linear and nonlinear elastic constants for planar QCs. All results are listed in Table 1.

From this table one can see that within linear phonon elasticity icosahedral QCs behave like isotropic media, while their nonlinear phonon elastic properties can manifest the anisotropic structure. Compared with icosahedral QCs all planar QCs have isotropic elastic properties within both linear and nonlinear phonon domain.

Since QCs have additional phason degree of freedom, its contribution to the elastic property should be considered. We analyze the phason influence below.

As we know, a vector $\mathbf{v}$ by its definition transforms according to

$$\mathbf{v}'=\mathbf{D}(\mathbf{R})\mathbf{v}\qquad(\mathbf{R}\in\mathbf{G}),\qquad(13)$$

or in the component form

$$v_{i'}=D_{i'k}v_k,\qquad(14)$$

where $D_{i'k}$ is the element of the vector representation matrix $\mathbf{D}(\mathbf{R})$ and G is the symmetry group. A tensor $\mathbf{T}$ transforms into

$$T_{m'n'\cdots l'}=\underbrace{D_{m'i}D_{n'j}\cdots D_{l'k}}_{r}T_{ij\cdots k},\qquad(15)$$

which is called a rth-rank tensor. Here it is worth noting that this transformation is applicable only to the physical space V_E but not to the perpendicular space V_I. Thus for the phonon displacement, the rank of tensor obviously equals to the number of its subscripts. However, the phason displacement, which is in the perpendicular space, does not transform under the vector representation but under a relevant irreducible representation. So it should have another tensor property, in other words, it is a tensor of high rank.

For instance, the vector irreducible representation associated with the phonon displacement is $\Gamma_1+\Gamma_3$, while the irreducible representation associated with the phason displacement is Γ_5 for octagonal quasicrystal [20]. Since we focus on the quasiperiodic plane, the part of the vector irreducible representation that we interest in is only Γ_3. Then the direct product $\Gamma_3\times\Gamma_3$ can be written as

$$\Gamma_3\times\Gamma_3=2\Gamma_1+\Gamma_4.\qquad(16)$$

Noticing that Γ_5 firstly enters the decomposition of the triple product

$$\Gamma_3\times\Gamma_3\times\Gamma_3=\Gamma_3\times(2\Gamma_1+\Gamma_4)=3\Gamma_3+\Gamma_5,\qquad(17)$$

one can consider the phason displacement w_α as a tensor of rank 3, and the phason strain $W_{\alpha i}$ is a tensor of rank 4. Thus, for octagonal quasicrystals the corresponding rank of C_{ijkl}, $K_{\alpha i\beta j}$, $R_{ij\alpha k}$, C_{ijklmn}, $K_{\alpha i\beta j\gamma k}$, $R^{(1)}_{ijkl\alpha m}$, $R^{(2)}_{ij\alpha k\beta l}$ are 4, 8, 6, 6, 12, 8, 10, respectively. In the same manner, the rank of these elastic constants for all QCs (see Table 2) can easily calculated.

As mentioned above, a macroscopic property is isotropic unless it is described by a tensor with rank $r\geqslant 8$ for octagonal QCs. Thus, it can be concluded that the nonlinear phonon elastic properties of octagonal QCs are isotropic as well, which coincides with the result given in Table 1. However, their third-order elastic properties due to phonon-phason coupling may be expected to be anisotropic. The results of whether the elastic properties of QCs are anisotropic or not are listed in Table 3.

3 Thermal expansion, piezoelectricity and piezoresistance of quasicrystals

Besides the elastic tensor, it must be considered the contributions of phason fields for thermal expansion tensors, piezoelectric tensors, piezoresistance tensors.

Table 2. The rank of second- and third-order elastic tensors for QCs.

	C_{ijkl}	$K_{\alpha i\beta j}$	$R_{ij\alpha k}$	C_{ijklmn}	$K_{\alpha i\beta j\gamma k}$	$R^{(1)}_{ijkl\alpha m}$	$R^{(2)}_{ij\alpha k\beta l}$
Icosahedral QCs	4	8	6	6	12	8	10
Decagonal QCs	4	8	6	6	12	8	10
Octagonal QCs	4	8	6	6	12	8	10
Dodecagonal QCs	4	12	8	6	18	10	14

Table 3. Isotropic or anisotropic elastic constants for QCs.

	C_{ijkl}	$K_{\alpha i\beta j}$	$R_{ij\alpha k}$	C_{ijklmn}	$K_{\alpha i\beta j\gamma k}$	$R^{(1)}_{ijkl\alpha m}$	$R^{(2)}_{ij\alpha k\beta l}$
Icosahedral QCs	I	A	A	A	A	A	A
Decagonal QCs	I	I	I	I	A	I	A
Octagonal QCs	I	A	I	I	A	A	A
Dodecagonal QCs	I	A	I	I	A	I	A

Note: "I" for isotropic, "A" for Anisotropic.

The thermoelastic effect can be represented as the change of strains induced by the temperature:

$$E_{ij} = \alpha^{(1)}_{ij}\theta, W_{\alpha i} = \alpha^{(2)}_{\alpha i}\theta, \tag{18}$$

where θ is temperature, $\alpha^{(1)}_{ij}$ and $\alpha^{(2)}_{\alpha i}$ the thermal expansion coefficients, and the superscripts (1) and (2) denote the components associated with the phonon field and phason field. Furthermore, from the character orthogonality theorem we can infer that $\alpha^{(2)}_{\alpha i} = 0$ for all QCs [20].

It is well-known that there are two kinds of strain fields: the phonon strain E_{ij} and the phason strain $W_{\alpha i}$, and two kinds of stress fields: the phonon stress T_{ij} and the phason stress $H_{\alpha i}$ in the case of QCs.

The relation between the electric displacement vector D_i and stresses T_{ij}, $H_{\alpha j}$ is

$$D_i = d^{(1)}_{ijk}T_{jk} + d^{(2)}_{i\alpha j}H_{\alpha j}, \tag{19}$$

where $d^{(1)}_{ijk}$ and $d^{(2)}_{i\alpha j}$ are the piezoelectric tensors associated with the phonon field and the phason field, respectively. And for icosahedral quasicrystals, $d^{(1)}_{ijk} = d^{(2)}_{i\alpha j} = 0$.

The piezoresistance effect can be described as a change of the resistivity tensor ρ_{ij} under the action of stressed T_{mn} and $H_{\alpha n}$:

$$\varepsilon_i = (\rho^0_{il} + P^{(1)}_{ilmn}T_{mn} + P^{(2)}_{il\alpha n}H_{\alpha n})J_l = (\rho^0_{il} + \rho^1_{il} + \rho^2_{il})J_l, \tag{20}$$

where ε is the electric field intensity, $\mathbf{J}$ is the current density, ρ^0_{il} is the resistivity tensor of a QC in the absence of mechanical stresses, ρ^1_{il} and ρ^2_{il} are the resistivity tensors induced by phonon and phason stresses, respectively, and $P^{(1)}_{ilmn}$ and $P^{(2)}_{il\alpha n}$ are the piezoresistance tensors.

In the same manner, we can estimate with the aid of Hermann's Theorem whether these physical properties of QCs are anisotropic or not. The results are given in Table 4. From this table it follows that: (1) thermal expansion are isotropic for all QCs; (2) piezoelectric and piezoresistance properties are isotropic for planar decagonal, octagonal and dodecagonal quasicrystals; (3) piezoresistance properties due to phonon field are still isotropic for icosahedral quasicrystals, while those due to phason field are expected to be anisotropic.

Table 4. Isotropic or anisotropic thermal expansion, piezoelectric and piezoresistance constants for QCs.

	$\alpha^{(1)}_{ij}$	$\alpha^{(2)}_{\alpha i}$	$d^{(1)}_{ijk}$	$d^{(2)}_{i\alpha j}$	$P^{(1)}_{ilmn}$	$P^{(2)}_{il\alpha n}$
Icosahedral QCs	I	–	–	–	I	A
Decagonal QCs	I	–	I	I	I	I
Octagonal QCs	I	–	I	I	I	I
Dodecagonal QCs	I	–	I	I	I	I

4 Conclusion

In this paper we investigate some physical properties such as linear and nonlinear elasticity, thermal expansion, piezoelectricity, and piezoresistance to reveal the quasicrystal anisotropy. According to Hermann's Theorem the minimal tensorial rank which is necessary to reveal the anisotropy of a symmetry is related to the order of the symmetry rotation. It follows that a macroscopic property is isotropic unless it is described by a tensor with rank $r \geqslant N$, which N is the order of the QC symmetry rotation. As we know, the phonon displacement transforms under the vector representation, whereas the phason displacement transforms under another irreducible representation. It is easy to find out the tensorial rank of the phason displacements for QCs and the corresponding rank of physical properties.

Based on these facts one can estimate whether the physical properties are anisotropic or not. The main results are:

For the elastic properties of quasicrystals (1) linear phonon elastic properties of all quasicrystals are isotropic; (2) nonlinear phonon elastic properties of icosahedral quasicrystals are anisotropic; (3) nonlinear phonon elastic properties of decagonal, octagonal and dodecagonal quasicrystals are still isotropic; (4) the anisotropic elasticities of decagonal, octagonal and dodecagonal quasicrystals due to phonon-phason coupling in the nonlinear elastic domain may be revealed in experiments.

For other physical properties (1) thermal expansion are isotropic for all QCs; (2) piezoelectricity and piezoresistance are isotropic for decagonal, octagonal and dodecagonal quasicrystals; (3) piezoresistance due to phonon field are still isotropic, but those due to phason field are expected to be anisotropic for icosahedral quasicrystals.

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