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Piezoelectricity in quasicrystals: A group-theoretical study

K RAMA MOHANA RAO[1], P HEMAGIRI RAO[2] and B S K CHAITANYA[3]
[1]Department of Applied Mathematics, Andhra University, Visakapatnam 530 003, India
[2]Department of Computer Applications, Vasavi College of Engineering, Hyderabad 500 031, India
[3]Department of Mathematics, DNR College (P.G. Courses), Bhimavaram 534 202, India
E-mail: hemagiriraop@yahoo.co.in

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Abstract. Group-theoretical methods have been accepted as exact and reliable tools in studying the physical properties of crystals and quasicrystalline materials. By group representation theory, the maximum number of non-vanishing and independent second-order piezoelectric coefficients required by the seven pentagonal and two icosahedral point groups – that describe the quasicrystal symmetry groups in two and three dimensions – is determined. The schemes of non-vanishing and independent second-order piezoelectric tensor components needed by the nine point groups with five-fold rotations are identified and tabulated employing a compact notation. The results of this group-theoretical study are briefly discussed.

Keywords. Quasicrystals; pentagonal and icosahedral point groups; piezoelectricity; non-vanishing and independent tensor coefficients; irreducible representations; composition series.

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1. Introduction

Ever since the discovery of quasicrystals, there has been a tremendous burst of theoretical and experimental research activity on the structure determination and physical property studies of these quasiperiodic materials. With the discovery of aluminum–copper–lithium (Al_6CuLi_3) icosahedral phase [1] forming an equilibrium compound with single grain approaching 1 mm size, followed by Al–Fe–Cu and Al–Pd–Mn systems [2,3] forming stable phases of perfect (phason-less) quasicrystals, the experimental situation was significantly improved. Thermodynamically stable, perfect quasicrystals such as Al–Cu–Li, Al–Cu–Ru [4] and Al–Pd–Cr–Fe [5] identified during this period, provided much information about the structural and physical property studies. These new materials posses a high degree of structural perfection – compared to that found in the periodic alloys – and provide better understanding of the microstructure of quasicrystals. They also provide an opportunity for studying the dislocations, hydrodynamic theory, elasticity [6–8] and electronic properties such as resistivity, magnetoresistance and Hall effect [9].

The discovery of optically active and transparent quasicrystals such as rare earth pyrogerminate (RPG) quasicrystal ($R_2Ge_2O_7$) and thulium pyrogerminate (TmPG) one with a unique crystal field potential of $\overline{10}m2$ (D_{5h}) site symmetry [8] have opened new avenues for group-theoretical physicists to study some magnetic and physical properties and derive schemes of non-vanishing and independent tensor components for the seven pentagonal and two icosahedral point groups. In analogy with the case of crystals, the classical group-theoretical methods were extended to quasicrystals by several investigators to unravel various physical properties. For instance, Brandmuller and Claus [10,11] have calculated the irreducible tensors of rank 1–4 (without intrinsic symmetries) for all the irreducible representations (IRs) of the pentagonal and icosahedral point groups which are useful for evaluating the property tensor components and interpreting Raman and hyper-Raman scattering. Jiang-Yi-Jian *et al* [12] derived the first-order photoelastic, piezoelectric and Brillouin tensor coefficients and second-order elastic tensor coefficients. Motivated by these findings, Rama Mohana Rao and Hemagiri Rao obtained the non-vanishing and independent piezomagnetic, pyromagnetic and magnetoelectric polarizability tensor coefficients [13]; third-order elastic coefficients and second-order piezomagnetic tensor coefficients [14] and later the second-order photoelastic coefficients [15] for these nine point groups with five-fold rotations. Following these studies, Wenge Yang *et al* [16] studied the thermodynamics of equilibrium properties and obtained the pyroelectric, electrocaloric and first-order piezoelectric coefficients and showed that the results obtained by them were in agreement with those of the earlier investigators [10,12] in respect of the first-order piezoelectric coefficients. Recently, Xiang Zhou *et al* [17] investigated the piezoresistance properties of quasicrystals due to phonon and phason stresses and derived the number of independent components of the piezoresistivity tensor for the three-dimensional icosahedral and two-dimensional quasicrystals. The references cited here indicate the spurt of recent activity in this area of research.

The present paper is concerned with the group-theoretical study of second-order piezoelectricity in quasicrystals. In §2, the enumeration of the number of non-vanishing and independent second-order piezoelectric coefficients needed for the nine quasicrystalline classes with five-fold rotations, $5(C_5)$, $\overline{5}(S_{10})$, $\overline{10}(C_{5h})$, $\overline{10}m2(D_{5h})$, $52(D_5)$, $5m(C_{5v})$, $\overline{5}m2(D_{5d})$; $235(I)$, $\frac{2}{m}\overline{35}(I_h)$ is carried out with the help of one-dimensional irreducible representation (IR) of the factor groups G_i/G_{i+1} contained in the composition series, that exist among these nine-point groups [13]. In the section that follows, the non-vanishing and independent components of second-order piezoelectric tensor are evaluated and listed, employing a compact notation. The paper ends with a brief discussion of the results and a few concluding remarks.

2. Enumeration of second-order piezoelectric coefficients

It is well-known that piezoelectricity is a physical property that connects two physical quantities, namely, stress and electric polarization. If $P_i, i = 1, 2, 3$ denote the components of electric polarization moment per unit volume and τ_{jk} stand for the components of a symmetric second-rank stress tensor, then we may write

$$P_i = d_{ijk}\tau_{jk};\ i, j, k, = 1, 2, 3. \tag{2.1}$$

Here d_{ijk} are 18 in number and constitute a tensor of rank 3. The independent components of the first-order piezoelectric tensor for each of the nine aforesaid point groups were derived by Jiang Yi-Jain *et al* [12] and Wenge Yang *et al* [16].

The physical property of second-order piezoelectricity on the other hand represents the relation between the axial vector and fourth-order symmetric stress tensor. Hence, the second-order piezoelectric tensor d_{ipqrs} can be expressed as a product of vector and square of a symmetric tensor. As such it is a fifth rank tensor invariant with respect to an interchange of p with q or r with s as also with the pairs pq and rs. When these tensor components are written in three suffix notation, the independent components are those d_{ijk} whose suffixes take values $1 \le i \le 3$ and $1 \le j, k \le 6$ with $j \le k$. Hence, its compound character $\chi^{\Gamma}(\mathrm{R})$ can be written [18] as

$$\chi^{\Gamma}(R_\phi) = (2c \pm 1)(16c^4 \pm 8c^3 - 4c^2 + 1). \tag{2.2}$$

In eq. (2.2), c stands for the cosine of the angle of rotation ϕ represented by the symmetry operation R of the point group under consideration. The positive or negative sign is to be taken according to whether the symmetry operation R under consideration is a pure rotation or rotation reflection through an angle ϕ.

The maximum number (n_i) of non-vanishing and independent second-order piezoelectric coefficients of the property tensor under consideration are determined here with respect to each of the nine quasicrystalline classes with five-fold rotations by considering (i) the total symmetric IR of the factor groups G_i/G_{i+1} contained in the composition series, $G = G_0 \supset G_1 \supset G_2 \supset \cdots \supset G_i \supset G_{i+1} \supset \cdots \supset 1$ that exist among the seven pentagonal and two icosahedral point groups, (ii) the definition of the character of the coset for any physical property [13], (iii) the computed character $\chi^{\Gamma}(R_\phi)$ corresponding to the symmetry operation R_ϕ in the representation provided by the second-order piezoelectric property and (iv) the well-known formula [19]:

$$n_i = \frac{1}{N}\sum_{\rho} h_\rho \chi_\rho^{(\Gamma_i)} \chi_\rho^{(\Gamma)} \tag{2.3}$$

with the usual notation. The results obtained are presented in table 1.

3. Evaluation of the second-order piezoelectric tensor coefficients

The non-vanishing and independent second-order piezoelectric tensor components are evaluated with respect to each of the nine quasicrystalline classes in this section using the tensor transformation law:

$$d^1_{ijklm} = a_{ip}a_{jq}a_{kr}a_{ls}a_{mt}d_{pqrst}, i, j, k, l, m = 1, 2, 3 \tag{3.1}$$

Table 1. Number (n_i) of independent constants required to describe the second-order piezoelectricity by the seven pentagonal and two icosahedral point groups.

Pentagonal/ icosahedral point groups	Number of independent constants required to describe second-order piezoelectricity
5	13
$\overline{5}$	0
$\overline{10}$	2
$\overline{10}m2$	1
52	4
$5m$	9
$\overline{5}2m$	0
235	0
$\frac{2}{m}\overline{35}$	0

and solving the equations that arise when imposing the condition that the tensors are invariant under the symmetry transformations of the point group. The complexity in computation is reduced by considering the composition series that exists among the nine point groups and evaluating the non-vanishing components of the group G_i in the series, from those of the independent non-vanishing components of the maximal normal subgroup G_{i+1} by the application of the appropriate generator(s) g_i that generate G_i from G_{i+1}. The simplified procedure is illustrated here by considering the series:

$$\overline{10}m2 \supset \overline{10} \supset 5 \supset 1 \tag{i}$$

and

$$\tfrac{2}{m}\overline{35} \supset 235 \supset 1. \tag{ii}$$

Point group 1 requires 63 independent second-order piezoelectric coefficients. The coefficients needed by point group 5 are obtained from those of point group 1 (C_1) by the application of the generating symmetry operation C_{5z}. Upon solving the obtained transformation equations we find a scheme with 13 independent coefficients for point group 5 (C_5). Application of the generating symmetry operation σ_h on the scheme of 5 in conjuction with eq. (3.1) gives the two independent coefficients required by point group $\overline{10}(C_{5h})$. Similarly, by operating C_2 on the coefficients obtained for the point group $\overline{10}$, we find that one independent coefficient is necessary for point group $\overline{10}m2$ (D_{5h}).

By identifying the appropriate generators and adopting the same procedure for series (ii), we find that both the icosahedral point groups 235(I) and $\frac{2}{m}\overline{35}(I_h)$ require zero second-order piezoelectric coefficients. The non-vanishing and independent schemes of second-order piezoelectric coefficients derived for all the nine considered point groups are listed in table 2 employing the compact notation [15,20]. To avoid the excessive use of symbols, the letter d is omitted in all the entries of this table. For example, an entry 1 1 1 in the table stands for $d_{1\,1\,1}$.

Table 2. Second-order piezoelectric coefficients required for the seven pentagonal and two icosahedral point groups.

Quasicrystal class/ constants	5 (13)	52 (4)	$5m$ (9)	$\bar{5}$, $\bar{5}2m$ (0)	$\overline{10}$ (2)	$\overline{10}m2$ (1)	I, I_h (0)
111	111	111	111	0	111	111	0
112	−111	−111	−111	0	−111	−111	0
113	0	0	0	0	0	0	0
114	114	114	0	0	0	0	0
115	115	0	115	0	0	0	0
116	116	0	0	0	116	0	0
122	111	111	111	0	111	111	0
123	0	0	0	0	0	0	0
124	124	124	0	0	0	0	0
125	125	0	125	0	0	0	0
126	−116	0	0	0	−116	0	0
133	0	0	0	0	0	0	0
134	134	134	0	0	0	0	0
135	135	0	135	0	0	0	0
136	0	0	0	0	0	0	0
144	0	0	0	0	0	0	0
145	0	0	0	0	0	0	0
146	a	0	a	0	0	0	0
155	0	0	0	0	0	0	0
156	b	b	0	0	0	0	0
166	−111	−111	−111	0	−111	−111	0
211	116	0	0	0	116	0	0
212	−116	0	0	0	−116	0	0
213	0	0	0	0	0	0	0
214	125	0	125	0	0	0	0
215	−124	−124	0	0	0	0	0
216	−111	−111	−111	0	−111	−111	0
222	116	0	0	0	116	0	0
223	0	0	0	0	0	0	0
224	115	0	115	0	0	0	0
225	−114	−114	0	0	0	0	0
226	111	111	111	0	111	111	0
233	0	0	0	0	0	0	0
234	135	0	135	0	0	0	0
235	−134	−134	0	0	0	0	0
236	0	0	0	0	0	0	0
244	0	0	0	0	0	0	0
245	0	0	0	0	0	0	0
246	$-b$	$-b$	0	0	0	0	0
255	0	0	0	0	0	0	0
256	a	0	a	0	0	0	0
266	−116	0	0	0	−116	0	0
311	311	0	311	0	0	0	0

Table 2. Contd...

Quasicrystal class/ constants	5 (13)	52 (4)	$5m$ (9)	$\bar{5}$, $\bar{5}2m$ (0)	$\overline{10}$ (2)	$\overline{10}m2$ (1)	I, I_h (0)
3 1 2	3 1 2	0	3 1 2	0	0	0	0
3 1 3	3 1 3	0	3 1 3	0	0	0	0
3 1 4	0	0	0	0	0	0	0
3 1 5	0	0	0	0	0	0	0
3 1 6	0	0	0	0	0	0	0
3 2 2	3 1 1	0	3 1 1	0	0	0	0
3 2 3	3 1 3	0	3 1 3	0	0	0	0
3 2 4	0	0	0	0	0	0	0
3 2 5	0	0	0	0	0	0	0
3 2 6	0	0	0	0	0	0	0
3 3 3	3 3 3	0	3 3 3	0	0	0	0
3 3 4	0	0	0	0	0	0	0
3 3 5	0	0	0	0	0	0	0
3 3 6	0	0	0	0	0	0	0
3 4 4	3 4 4	0	3 4 4	0	0	0	0
3 4 5	0	0	0	0	0	0	0
3 4 6	0	0	0	0	0	0	0
3 5 5	3 4 4	0	3 4 4	0	0	0	0
3 5 6	0	0	0	0	0	0	0
3 6 6	c	0	c	0	0	0	0

$a = 1/2(d_{115} - d_{125})$; $b = 1/2(d_{124} - d_{114})$; $c = 1/2(d_{311} - d_{312})$.

4. Conclusions

The simple and elegant group-theoretical method adopted in this paper for enumerating and evaluating the non-vanishing and independent second-order piezoelectric coefficients avoids considering each one of the nine quasicrystalline classes separately. The tensor components of a quasicrystalline class G_i are obtained from those of the components of a maximal normal subgroup G_{i+1} in the considered series by just applying the appropriate generator(s) g_i that generates G_i from G_{i+1}.

It can be seen that the maximum number of independent piezoelectric coefficients needed by the quasicrystalline classes increase with the increasing order of the effect. It was found that for the first-order effect the maximum number was only four [12] whereas for the second-order effect we find that it is thirteen, for the nine-point groups under consideration.

The number of independent first-order piezoelectric coefficients for the icosahedral classes was found to be zero [12]. It is interesting to note here that for the icosahedral classes, this number for the second-order effect is also zero – indicating that the icosahedral classes 235 and $\frac{2}{m}\overline{35}$ do not exhibit piezoelectric behavior of known order.

Most scientists working in this area are unanimous in their opinion that quasicrystalline materials with icosahedral and other quasiperiodic symmetries represent a new phase of matter with possibly unique physical properties which one has to identify and understand. These materials are generally hard ($Hv > 500$) but ductile in certain conditions. They are good conductors of heat, are thermally stable and can be chemically adjusted to meet corrosion problems. Accordingly they are mild ware applications, ranging from non-stick frying pans to a self-lubricated combustion chamber in heat engines [21]. Since quasicrystalline nature is observed and identified in several composite materials, experimental evaluation/verification of the theoretical results is naturally expected in these materials. But to-date only a limited experimental work in the study of physical properties has been carried out. As Cheng Zheng Hu *et al* [22] observed, 'some preliminary investigations have been made in this field' (and) 'most of the physical properties predicted in QCs still remain to be confirmed experimentally'.

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