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Elasticity and piezomagnetism in pentagonal and icosahedral quasi-crystals: a group-theoretical study

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Abstract. The maximum number of independent third-order elastic and second-order piezomagnetic constants required by the seven pentagonal point groups 5, $\bar{5}$, $\bar{1}\bar{0}$, 52, $5m$, $\bar{5}2m$, and $\bar{1}\bar{0}m2$ and the two icosahedral point groups 235 and $2/m\bar{3}\bar{5}$ that represent the symmetries of quasi-crystals in two and three dimensions are obtained in this paper. The non-vanishing and independent tensor components needed by each of these nine point groups with fivefold rotations are identified and tabulated. The results of this group-theoretical study are briefly discussed and summarized.

1. Introduction

Ever since Schechtman *et al* (1984) in an exciting experiment on manganese–aluminium alloy ($Al_{86}Mn_{14}$) observed a diffraction spectrum with fivefold symmetry, inconsistent with the usual lattice translation for periodic crystals, there has been tremendous progress in theoretical and experimental research activity (see, e.g., Levine and Steinhardt (1984), Gratias and Michel (1986) and Mackay (1987)) towards understanding these structures to determine whether the discovered alloys are indeed quasi-crystals. This study culminated in the detection of many alloys exhibiting icosahedral phases similar to Al_4Mn; the rapid solidification of an aluminium–copper–lithium quasi-crystal (Al_6CuLi_3) by Dubost *et al* (1986) and Bartges *et al* (1987), the optically active transparent rare-earth pyrogerminate (RPG) quasi-crystal ($R_2Ge_2O_7$) and thulium pyrogerminate (TmPG) with a unique crystal-field potential of $\bar{1}\bar{0}m2$ (D_{5h}) site symmetry by Sen Gupta *et al* (1988) and the latest quasi-crystals with simple metals such as Al–Cu–Fe, Al–Mg–Zn (Carlsson 1991) and the Al–Cu–Ru quasi-crystals reported by Vincenzo (1989) are only a few interesting recent additions to the class of quasi-crystalline materials exhibiting fivefold rotational symmetry. The quasi-crystal model thus remains a leading explanation for these new alloys.

Experimental measurements of the physical properties of the icosahedral phases of Al–Mn and other related alloys have been hampered by the fact that the grain size has been very small (less than 50 μm). The recent discovery of new alloys with much larger single grains in Al_6CuLi_3 by Sainfort and Silcock (1985), Dubost *et al* (1986) and Bartges *et al* (1987) and the rapid solidification of the $Al_{63}Cu_{25}Fe_{12}$ quasi-crystal by Biggs *et al* (1990) promise improvement in this regard. Although definitive (experimental) results on the physical properties of quasi-crystals have not been published to date—so far as the present authors know—extensive theoretical progress has been made in recent years on

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the stability, the hydrodynamic theory, and the elastic, photo-elastic, piezoelectric and a few magnetic properties of quasi-crystals†: Levine *et al* (1985) developed theories for elasticity and dislocation defects in two-dimensional (2D) pentagonal and three-dimensional (3D) icosahedral quasi-crystals. Bak (1985) studied the symmetry, stability and elastic properties phenomenologically of icosahedral diffraction patterns and obtained the acoustic phonon and phason modes. Sen Gupta *et al* (1988) worked out the crystal-field effects on the magnetic properties of TmPG and obtained expressions for magnetic susceptibility tensors under a crystal field of D_{5h} symmetry. Brandmuller and Clauss (1988a, b) 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 interpreting Raman and hyper-Raman scattering. Jiang Yi-Jian *et al* (1990) have obtained the first order piezoelectric and photoelastic tensor coefficients and second-order elastic tensor coefficients for all the nine point groups with fivefold rotations and on the basis of these results derived the Brillouin tensors with such symmetries. In a recent communication, the present authors (Rama Mohana Rao and Hemagiri Rao 1992) with the employment of group-theoretical methods have obtained the maximum number of non-vanishing and independent piezomagnetic, pyromagnetic and magnetoelectric polarizability constants required for all the point groups with fivefold rotations and determined the non-vanishing and independent tensor coefficients corresponding to the magnetic properties considered.

The purpose of the present paper is twofold, namely

(i) to obtain through group-theoretical methods the number of non-vanishing and independent third-order elastic coefficients and second-order piezomagnetic coefficients for the seven pentagonal point groups $5(C_5)$, $\bar{5}(S_{10})$, $\bar{10}(C_{5h})$, $\bar{10}m2(D_{5h})$, $52(D_5)$, $5m(C_{5v})$ and $\bar{5}2m(D_{5d})$ and the two icosahedral point groups 235 (I) and $2/m\bar{3}\bar{5}(I_h)$ which are the quasi-crystals' symmetry groups in two and three dimensions and

(ii) to identify the surviving tensor components corresponding to these physical properties for the point groups considered.

This paper is organized as follows. In section 2, we discuss briefly the phenomenon of third-order elasticity and second-order piezomagnetism and indicate the structure for their compound character, expressed as the product of the characters of the quantities involved in defining these two physical properties. In section 3, the procedure for obtaining the maximum number of non-vanishing as well as independent third-order elastic and second-order piezomagnetic constants in respect of all the nine quasi-crystalline classes is indicated and the independent schemes so obtained of the non-vanishing tensor coefficients are tabulated. Finally the results obtained in this work are briefly discussed in section 4. The notation for the coefficients given in tables 2 and 3 is explained in the appendix.

2. Third-order elastic and second-order piezomagnetic coefficients: a resumé

2.1. Third-order elasticity

In the classical theory of elasticity, the strains are assumed to be infinitesimal and the resulting strain energy function is a homogeneous quadratic function of the strains. However, if the strains are not infinitesimal, then terms of third and higher degree in the strains enter into the strain energy function (Kaplan 1931). If the initial energy and the initial cubic

† A comprehensive account of the elasticity of crystals and quasi-crystals has been provided by Jaric (1986).

Table 1. Number n_i of independent constants required to describe the third-order elasticity and second-order piezomagnetism by the seven pentagonal and two icosahedral quasi-crystalline classes.

Pentagonal or icosahedral point group	Number of constants required to describe: Third-order elasticity	Second-order piezomagnetism
5	12	13
$\bar{5}$	12	13
$\overline{10}$	10	11
$\overline{10}m2$	9	3
52	10	4
$5m$	10	4
$\bar{5}2m$	10	4
235	4	0
$2/m\bar{3}\bar{5}$	4	0

dilation of the body are zero, the expression for the energy deformation of a body can be expressed as

$$\phi = \tfrac{1}{2}C_{ijkl}\eta_{ij}\eta_{kl} + C_{ijklmn}\eta_{ij}\eta_{kl}\eta_{mn} + \cdots. \tag{2.1}$$

In equation (2.1), the suffixes take the values 1, 2, 3 and the summation convention is implied with respect to a repeated suffix. Whereas the C_{ijkl} are the elastic stiffness coefficients which form a fourth-rank tensor, the C_{ijklmn} are known as the third-order elastic coefficients. The latter form a sixth-rank tensor containing 729 components. The intrinsic symmetry reduces the maximum number of independent components permissible to 56 (table A1 of the appendix). Birch (1947) derived the schemes of independent coefficients for all the classes of cubic crystals. Bhagavantam and Suryanarayana (1947, 1949) through a group-theoretical method obtained the number of independent third-order coefficients in each crystal class and in fact corrected Birch's result for one of the cubic classes. Jahn (1949) independently confirmed the results of Bhagavantam and Suryanarayana and extended the calculations to include isotropic materials, and Hearmon (1953) tackled the general problem of third-order elastic coefficients of crystals independently.

Since the physical property of third-order elasticity represents the relation between the symmetric tensor and the square of the symmetric tensor, the compound character $\chi^{(\Gamma)}$ representing this property is given by

$$\chi_e^{(\Gamma)}(R_\phi) = 64c^6 \pm 32c^5 - 48c^4 \mp 8c^3 + 16c^2. \tag{2.2}$$

In equation (2.2), the positive or negative sign is to be taken according to whether the symmetry operation R_ϕ in question is a pure rotation or a rotation–reflection through an angle ϕ and $c = \cos\phi$.

2.2. *Second-order piezomagnetism*

It can be seen that the phenomenon of piezomagnetism is the appearance of a magnetic moment M (M_i; $i = 1, 2, 3$) by the application of stress σ and the piezomagnetic coefficients C_{ijk} of the first order are studied from the governing relation

$$M_i = \sum_j \sum_k C_{ijk}\sigma_{jk} \qquad i, j, k = 1, 2, 3. \tag{2.3}$$

Table 2. The non-vanishing and independent components of the third-order elastic tensor for the seven pentagonal and two icosahedral point groups.

Point group	Non-vanishing and independent third-order elastic tensor components						
$5, \bar{5}$	111	112	113	114	115	0	
		112	123	−114	−115	0	
			133	0	0	0	
				144	145	−115	
					155	114	
						116	
		111	113	114	115	0	
			133	0	0	0	
				155	−145	115	
					144	−114	
						166	(12)
			333	0	0	0	
				344	0	0	
					344	0	
						366	
				0	0	145	
					0	456	
						−114	
					0	−145	
						−115	
						0	
with $166 = \frac{1}{4}(111 - 112)$; $366 = \frac{1}{2}(113 - 123)$; $456 = \frac{1}{2}(155 - 144)$							
$52, 5m, \bar{5}2m$	111	112	113	114	0	0	
		112	123	−114	0	0	
			133	0	0	0	
				144	0	0	
					155	114	
						166	
		111	113	114	0	0	
			133	0	0	0	
				155	0	0	(10)
					144	−114	
						166	
			333	0	0	0	
				344	0	0	
					344	0	
						366	
				0	0	0	
					0	456	
						−114	
					0	0	
						0	
						0	
with $166 = \frac{1}{4}(111 - 112)$; $366 = \frac{1}{2}(113 - 123)$; $456 = \frac{1}{2}(155 - 144)$							

In (2.3), M is an axial vector and σ a second-rank symmetric polar tensor. The non-vanishing as well as independent first-order piezomagnetic coefficients for the seven pentagonal and the two icosahedral point groups have already been derived in an earlier paper by the present authors (Rama Mohana Rao and Hemagiri Rao 1992).

Since the second-order piezomagnetism represents a relation between the axial vector

Table 2. (continued)

Point group	Non-vanishing and independent third-order elastic tensor components						
$\overline{10}$	111	112	113	0	0	0	
		112	123	0	0	0	
			133	0	0	0	
				144	145	0	
					155	0	
						166	
		111	113	0	0	0	
			133	0	0	0	
				155	−145	0	
					144	0	
						166	(10)
			333	0	0	0	
				344	0	0	
					344	0	
						366	
				0	0	145	
					0	456	
						0	
					0	−145	
						0	
						0	

with $166 = \frac{1}{4}(111 - 112)$; $366 = \frac{1}{2}(113 - 123)$; $456 = \frac{1}{2}(155 - 144)$

$\overline{10}m2$	111	112	113	0	0	0	
		112	123	0	0	0	
			133	0	0	0	
				144	0	0	
					155	0	
						166	
		111	113	0	0	0	
			133	0	0	0	
				155	0	0	
					144	0	(9)
						166	
			333	0	0	0	
				344	0	0	
					344	0	
						366	
				0	0	0	
					0	456	
						0	
					0	0	
						0	
						0	

with $166 = \frac{1}{4}(111 - 112)$; $366 = \frac{1}{2}(113 - 123)$; $456 = \frac{1}{2}(155 - 144)$

and the symmetrized stress (quadratic combination of stresses), the coefficients μ_i for the second-order piezomagnetic effect can be studied from

$$\mu_i = \sum_j \sum_k \sum_l \sum_m C_{ijklm} \sigma_{jk} \sigma_{lm} \tag{2.4}$$

with the indices taking the values 1, 2, 3. As the character of the axial vector is given by

Table 2. (continued)

Point group	Non-vanishing and independent third-order elastic tensor components						
235, $2/m\bar{3}\bar{5}$	111	112	113	114	0	0	
		112	123	−114	0	0	
			133	0	0	0	
				144	0	0	
					155	114	
						166	
		111	113	114	0	0	
			133	0	0	0	
				155	0	0	(4)
					144	−114	
						166	
			333	0	0	0	
				344	0	0	
					344	0	
						366	
				0	0	0	
					0	456	
						−114	
					0	0	
						0	
						0	

with $114 = 112 - 113$; $133 = 2(112) - 113$; $144 = 366 = \frac{1}{2}(113 - 123)$;
$155 = \frac{1}{4}[111 - 5(112) + 4(113)]$; $166 = \frac{1}{4}(111 - 112)$;
$333 = \frac{1}{4}[4(111) - 33(112) + 33(113)]$; $344 = \frac{1}{4}[111 + 3(112) - 4(113)]$;
$456 = \frac{1}{8}[111 - 5(112) + 2(113) + 2(123)]$

$1 \pm 2\cos\phi$ and that of the symmetrized stress by

$$16\cos^4\phi \pm 8\cos^3\phi - 4\cos^2\phi + 1$$

the compound character representing the second-order piezomagnetism can be expressed as

$$\chi_{\rm p}^{(\Gamma)}(R_\phi) = (1 \pm 2\cos\phi)(16\cos^4\phi \pm 8\cos^3\phi - 4\cos^2\phi + 1) \qquad (2.5)$$

with the understanding as before about the alternative signs when they occur.

3. Third-order elastic and second-order piezomagnetic coefficients

The group-theoretical method employed to obtain the desired coefficients is a straightforward application of the method used hitherto to analyse periodic crystals (Bhagavantam and Suryanarayana 1947, 1949, Hearmon 1953) and also quasi-crystals (Rama Mohana Rao and Hemagiri Rao 1992). The numbers n_i of the independent third-order elastic coefficients and second-order piezomagnetic coefficients for all the nine quasi-crystalline classes are obtained here with the help of the factor groups G_i/G_{i+1} contained in the composition series that exist among these nine point groups (Rama Mohana Rao and Hemagiri Rao 1992). The enumeration is done by considering

Table 3. Non-vanishing and independent components of the second-order piezomagnetic tensor for the seven pentagonal and two icosahedral point groups.

Point group	Non-vanishing and independent second-order piezomagnetic tensor						
5, $\bar{5}$	111	−111	0	114	115	116	
		111	0	124	125	−116	
			0	134	135	0	
				0	0	146	
					0	156	
						−111	
	116	−116	0	125	−124	−111	
		116	0	115	−114	111	
			0	135	−134	0	(13)
				0	0	−156	
					0	146	
						−116	
	311	312	313	0	0	0	
		311	313	0	0	0	
			333	0	0	0	
				344	0	0	
					344	0	
						366	
with $146 = \frac{1}{2}(115 - 125)$; $156 = \frac{1}{2}(124 - 114)$; $366 = \frac{1}{2}(311 - 312)$							
$\bar{10}$	0	0	0	114	115	0	
		0	0	124	125	0	
			0	134	135	0	
				0	0	146	
					0	156	
						0	
	0	0	0	125	−124	0	
		0	0	115	−114	0	
			0	135	−134	0	(11)
				0	0	−156	
					0	−146	
						0	
	311	312	313	0	0	0	
		311	313	0	0	0	
			333	0	0	0	
				344	0	0	
					344	0	
						366	
with $146 = \frac{1}{2}(115 - 125)$; $156 = \frac{1}{2}(124 - 114)$; $366 = \frac{1}{2}(311 - 312)$							

(i) the total symmetric IR of the factor group G_i/G_{i+1} in the composition series

$$G = G_0 \supset G_1 \supset \ldots \supset G_i \supset G_{i+1} \supset \ldots \supset G_s = E.$$

(ii) the character $\chi_e(R_\phi)$ and $\chi_p(R_\phi)$ corresponding to the element R_ϕ in the representation provided by the two physical properties considered (as given in equations (2.2) and (2.5)),

(iii) the definition of the character of a coset for any physical (magnetic) property (Rama Mohana Rao and Hemagiri Rao 1992) and

(iv) the well known formula (Bhagavantam and Venkatarayudu 1951)

Table 3. (continued)

Point group	Non-vanishing and independent second-order piezomagnetic tensor						
$\overline{10}m2$	0	0	0	114	0	0	
		0	0	124	0	0	
			0	134	0	0	
				0	0	0	
					0	156	
						0	
	0	0	0	0	−124	0	
		0	0	0	−114	0	(3)
			0	0	−134	0	
				0	0	156	
					0	0	
						0	
	0	0	0	0	0	0	
		0	0	0	0	0	
			0	0	0	0	
				0	0	0	
					0	0	
						0	
with $156 = \frac{1}{2}(124 - 114)$							
$52, 5m, \bar{5}2m$	111	−111	0	114	0	0	
		111	0	124	0	0	
			0	134	0	0	
				0	0	0	
					0	156	
						−111	
	0	0	0	0	−124	−111	
		0	0	0	−114	111	
			0	0	−134	0	(4)
				0	0	−156	
					0	0	
						0	
	0	0	0	0	0	0	
		0	0	0	0	0	
			0	0	0	0	
				0	0	0	
					0	0	
						0	
$235, 2/m\bar{3}\bar{5}$				0			(0)
with $156 = \frac{1}{2}(124 - 114)$							

$$n_i = \frac{1}{N} \sum_{\rho} h_\rho \chi_\rho^{(\Gamma_i)} \chi_\rho^{(\Gamma)} \tag{3.1}$$

with the usual notation.

The results obtained for all the seven pentagonal and two icosahedral point groups for the physical properties considered are presented in table 1.

The non-vanishing and independent tensor components in respect of each of the nine quasi-crystalline classes for the two physical properties are identified through a method similar to those of Birch (1947) and Hearmon (1953). We have calculated these components by solving the equations which arise when imposing the condition that the tensors are invariant under the elements of the point groups. The procedure is simplified here by considering the various point groups in the composition series and obtaining the non-vanishing components for G_i in the series, from those of the independent components of G_{i+1} by the application of the appropriate generator(s) g_i that generates G_i from G_{i+1}. The final results are provided in tables 2 and 3. The notation for the independent tensor components given in these two tables is explained in the appendix.

4. Discussion and summary of the results

The method adopted here for obtaining the elastic and piezomagnetic coefficients has the following advantage, namely it avoids considering each of the nine quasi-crystalline classes separately. The tensor components of a quasi-crystalline class G_i are obtained in a simple and elegant way from those of the components of a normal subgroup G_{i+1} in the series by just applying the appropriate generator(s) g_i that generates G_i from G_{i+1}.

It can be observed that, in respect of piezomagnetism, the nine classes divide into two categories:

(a) those classes which do not need piezomagnetic coefficients of any order (there are in fact two such classes: 235 and $2/m\bar{3}\bar{5}$) and

(b) those which need piezomagnetic coefficients but with different numbers for the first- and second-order effects.

Whereas for first-order piezomagnetism the seven pentagonal classes separate into two sets (5, $\bar{5}$, $\bar{1}0$; 52, $5m$, $\bar{1}0m2$, $\bar{5}2m$) requiring four coefficients and one coefficient, respectively (Rama Mohana Rao and Hemagiri Rao 1992), for the second order, these classes separate into four sets (5, $\bar{5}$; 52, $5m$, $\bar{5}2m$; $\bar{1}0$; $\bar{1}02m$) requiring 13, four, 11 and three coefficients, respectively.

In the case of elasticity, it can be observed that the elastic coefficients increase with increasing order of the effect. For the elastic coefficients of third order, the nine quasi-crystalline classes divide into four sets, whereas for second-order elasticity (Jiang Yi-Jian *et al* 1990) they divide into two sets, each of the seven pentagonal classes requiring five coefficients and the two icosahedral classes requiring two coefficients each. We find here that the seven pentagonal classes are split into three sets (5, $\bar{5}$; $\bar{1}0$, 52, $5m$, $\bar{5}2m$; $\bar{1}0m2$) requiring different numbers of coefficients (12, 10 and nine, respectively) and the two icosahedral classes forming a single set requiring four coefficients each. From table 1, one can see that there are only four independent third-order elastic constants for the icosahedral point groups, fewer than in any of the seven pentagonal classes and the 32 crystallographic point groups.

It is interesting that the maximum number of independent third-order elastic coefficients needed for the icosahedral class is four and the corresponding number for the isotropic medium is three, whereas the maximum number of elastic coefficients of second order for the icosahedral system and that for the isotropic medium are the same, namely two.

Not much experimental work seems to have been carried out on the study of physical properties of quasi-crystals as far as we know. We suggest that the group-theoretical work presented here on the two physical properties with respect to the pentagonal and icosahedral quasi-crystalline classes will be useful in further theoretical studies. Our results may also serve as valuable checks on the experimental studies pertaining to these physical properties for the class of new materials.

Table A1. The scheme showing the non-vanishing and independent components of the third-order elastic tensor for the point group 1.

Point group	Non-vanishing and independent tensor components						
1	111	112	113	114	115	116	
		122	123	124	125	126	
			133	134	135	136	
				144	145	146	
					155	156	
						166	
		222	223	224	225	226	
			233	234	235	236	
				244	245	246	(56)
					255	256	
						266	
			333	334	335	336	
				344	345	346	
					355	356	
						366	
				444	445	446	
					455	456	
						466	
					555	556	
						566	
						666	

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Appendix

The third-order elastic tensor C_{ijklmn} is invariant with respect to an interchange of one or the other of i, j or k, l or m, n, and also to an interchange of any of the three pairs ij, kl and mn with another. A sixth-rank tensor of three variables can in general have $3^6 = 729$ components. The intrinsic symmetry reduces the maximum number of independent components permissible to 56. This reduction can be obtained by writing the tensor components in the three-suffix notation as C_{ijk}. It is immediately seen that the independent components are those C_{ijk} which have suffixes taking values from 1 to 6 such that $i \leqslant j \leqslant k$. The maximum number of such coefficients is 56 and these are given in table A1.

The second-order piezomagnetic tensor d_{ijklm} is invariant with respect to interchange of j with k or l with m and also to an interchange of any of the two pairs jk and lm with another. A fifth-rank tensor of three variables can in general have $3^5 = 243$ components. The intrinsic symmetry reduces the maximum number of independent components permissible to 63. This reduction can be obtained by writing the tensor components in the three-suffix notation as d_{ijk}. It is immediately seen that the independent components are those d_{ijk} whose suffixes take values $1 \leqslant i \leqslant 3$ and $1 \leqslant j, k \leqslant 6$ with $j \leqslant k$. The number of such coefficients is 63 which are given in table A2.

Table A2. Scheme showing the non-vanishing and independent components of the second-order piezomagnetic tensor for the point group 1.

Point group	Non-vanishing and independent tensor components						
1	111	112	113	114	115	116	
		122	123	124	125	126	
			133	134	135	136	
				144	145	146	
					155	156	
						166	
	211	212	213	214	215	216	
		222	223	224	225	226	(63)
			233	234	235	236	
				244	245	246	
					255	256	
						266	
	311	312	313	314	315	316	
		322	323	324	325	326	
			333	334	335	336	
				344	345	346	
					355	356	
						366	

The complete list of third-order elastic coefficients C_{ijk} are given in the second column of table 2. Similarly the second-order piezomagnetic coefficients d_{ijk} are given in the second column of table 3. To avoid excessive use of the suffixes, the letter c is omitted from table 2 and d from table 3. Thus an entry such as 111 in table 2, for example, stands for c_{111}, and an entry such as 124 in table 3 stands for d_{124}.

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