research papers

Ferroic classifications extended to ferrotoroidic crystals

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Aizu's characterization of the 773 species of phase transitions by magnetization, polarization and strain is extended to include characterization by the recently observed toroidal moment. The resulting *distinction quadruplet* characterization is then used to classify the species into *sub-ensembles*, extending Schmid's concept of *ensembles* of species to include classification by toroidal moments. Tables are given of the distinction quadruplet characterization of each species and the species in each ensemble and sub-ensemble. The form of primary and secondary ferroic property tensors invariant under the 122 magnetic point groups have also been tabulated for use in determining the characterization of species and possible domain switching. In both cases, physical property tensors related to the toroidal moment are included.

1. Introduction

A ferroic crystal contains two or more equally stable *domain states*, volumes of the same homogeneous crystalline structure in different spatial orientations. These domains can coexist in a crystal and may be distinguished by the values of components of certain spontaneous macroscopic tensorial physical properties of the domains. Crystals in which the domains may be distinguished by spontaneous magnetization, polarization and strain are called *primary* ferroic crystals (Newnham, 1974; Wadhawan, 2000) and are individually referred to, respectively, as ferromagnetic, ferroelectric and ferroelastic ferroic crystals. The domains in these primary ferroic crystals can be switched by means of a magnetic field, an electric field, a mechanical stress or a combination of the three (Newnham & Cross, 1974a,b) and consequently ferroic crystals are of technological importance for, for example, memory storage and electric and magnetic switches.

Ferroic crystals arise in a phase transition from a paramagnetic prototypic state of symmetry $\mathbf{G1'}$, where $1'$ denotes time inversion and $\mathbf{G}$ a crystallographic point group, to a ferroic phase of lower symmetry $\mathbf{H}$. There are then $n = |\mathbf{G1'}|/|\mathbf{H}|$ domain states $\mathbf{S}_i$, $i = 1, 2, \ldots, n$, where $|\mathbf{G1'}|$ and $|\mathbf{H}|$ denote the number of elements in $\mathbf{G1'}$ and $\mathbf{H}$, respectively. Domain states refer to the bulk structures, with their specific orientations in space, of domains in a polydomain sample. Several disconnected domains can have the same domain state.

Aizu classified all possible ferroic phase transitions into 773 *species* (Aizu, 1970). Each species is given the symbol $\mathbf{G1'}F\mathbf{H}$ representing the transition between a paramagnetic phase of point-group symmetry $\mathbf{G1'}$ and a lower symmetry phase of symmetry $\mathbf{H}$. The F denotes ferroic. Aizu then characterized each species according to the ability of spontaneous magnetization, polarization and strain, individually, to distinguish among the domain states. Each spontaneous physical property was assigned one of three distinguishability types: able to distinguish among all, some or none of the domain states. Consequently, associated with each species is one of 27 ordered trios of distinguishability types which we call *distinction triplets*, *i.e.* the domain-state distinguishability by, respectively, spontaneous magnetization, polarization and strain.

Schmid (1999) classified the 773 species into 36 *ensembles*. Each ensemble consists of all species with the same distinction triplet. There are 36 and not 27 ensembles defined, as the zero spontaneous magnetization case was divided into two subtypes: the PDM, para- or diamagnetic, subtype where $\mathbf{H} = \mathbf{J1'}$ and the AFM, antiferromagnetic, subtype where $\mathbf{H}$ is a group which does not allow an invariant magnetization and which does not contain $1'$. The 773 species are all classified into 30 of these 36 ensembles with six ensembles containing no species.

A fourth type of primary ferroic crystals, a *ferrotoroidic crystal*, has been recently observed (Van Aken *et al.*, 2007), where the domains are distinguished by a toroidal moment (Gorbatsevich *et al.*, 1983; Schmid, 2001, 2003a). In §2, we extend Aizu's species characterization and Schmid's classification of species into ensembles to include ferrotoroidic crystals by including the domain-state distinguishability by a spontaneous toroidal moment. In Appendix *A*, for use in determining the species characterization and also possible domain switching, we have tabulated the tensor form of primary and secondary ferroic property tensors invariant under the 122 magnetic point groups.

Table 1
The four primary ferroics as bases for the four irreducible representations of the group $\bar{1}1' = \{1, \bar{1}, 1', \bar{1}'\}$ where $\bar{1}$ denotes spatial inversion and $1'$ time inversion.

1	$\bar{1}$	$1'$	$\bar{1}'$		Basis
1	1	1	1	ε_{ij}	Strain
1	−1	1	−1	P	Polarization
1	1	−1	−1	M	Magnetization
1	−1	−1	1	T	Toroidal moment

Table 2
Distinction quadruplet characterization of ferroic species T, M, P and ε_{ij} denote, respectively, toroidal moment, magnetization, polarization and strain; F, P, N and Z denote, respectively, full, partial, null and zero domain state distinction.

Species No.	Species		No.	T	M	P	ε_{ij}
					Distinction quadruplet		
114	$4221'F1$	$4_z2_x2_{xy}1'F1$	8×2	F	F	F	F
115	$4221'F1'$	$4_z2_x2_{xy}1'F1'$	8	Z	Z	F	F
116	$4221'F2(p)$	$4_z2_x2_{xy}1'F2_z$	4×2	P	P	P	F
117	$4221'F2(s)$	$4_z2_x2_{xy}1'F2_x$	4×2	P	P	F	F
118	$4221'F2'(p)$	$4_z2_x2_{xy}1'F2'_z$	4×2	F	F	P	F
119	$4221'F2'(s)$	$4_z2_x2_{xy}1'F2'_x$	4×2	F	F	F	F
120	$4221'F21'(p)$	$4_z2_x2_{xy}1'F2_z1'$	4	Z	Z	P	F
121	$4221'F21'(s)$	$4_z2_x2_{xy}1'F2_x1'$	4	Z	Z	F	F
122	$4221'F222$	$4_z2_x2_{xy}1'F2_x2_y2_z$	2×2	Z	Z	Z	F
123	$4221'F2'2'2(p)$	$4_z2_x2_{xy}1'F2'_x2'_y2_z$	2×2	P	P	Z	F
124	$4221'F2'2'2(s)$	$4_z2_x2_{xy}1'F2_x2'_y2'_z$	2×2	F	F	Z	F
125	$4221'F2221'$	$4_z2_x2_{xy}1'F2_x2_y2_z1'$	2	Z	Z	Z	F
126	$4221'F4$	$4_z2_x2_{xy}1'F4_z$	2×2	P	P	F	N
127	$4221'F4'$	$4_z2_x2_{xy}1'F4'_z$	2×2	Z	Z	F	N
128	$4221'F41'$	$4_z2_x2_{xy}1'F4_z1'$	2	Z	Z	F	N
129	$4221'F422$	$4_z2_x2_{xy}1'F4_z2_x2_{xy}$	1×2	Z	Z	Z	N
130	$4221'F42'2'$	$4_z2_x2_{xy}1'F4_z2'_x2'_{xy}$	1×2	F	F	Z	N
131	$4221'F4'2'2$	$4_z2_x2_{xy}1'F4'_z2'_x2_{xy}$	1×2	Z	Z	Z	N

2. Extension to ferrotoroidic crystals

Aizu's characterization of species is an example of *tensor distinction* (Litvin, 1999; see also Weiglhofer & Lakhtakia, 2003). In a phase transition associated with a species **G1′***F***H**, the specific forms of a physical property T in each of the n domain states S_i, $i = 1, 2, \ldots, n$, are given in a single coordinate system and denoted by T_i, $i = 1, 2, \ldots, n$. The tensor T_1 is invariant under the group **H**, the symmetry group of domain S_1. The tensors T_j, $j = 2, 3, \ldots, n$, in the remaining domains are given by $g_jT_1 = T_j$, $j = 2, 3, \ldots, n$, where the g_j, $j = 2, 3, \ldots, n$, are coset representatives of the coset decomposition of **G1′** with respect to **H**: **G1′** = **H** + g_2**H** + g_3**H** + $\ldots$ + g_n**H**. If the set of tensors T_i, $i = 1, 2, \ldots, n$, are all distinct, Aizu characterized the physical property T as *full* if some but not all are distinct, as *partial*, and with the notation '$\ldots$' if the set of tensors are all identical. Each species was characterized by three physical properties T, spontaneous magnetization, polarization and strain.

To extend the characterization of the 773 species to include ferrotoroidic crystals, we give in Table 1 the characteristic

transformation properties of a toroidal moment, along with that of the other three primary ferroics, under elements of the group $\bar{1}1' = \{1, \bar{1}, 1', \bar{1}'\}$, where $\bar{1}$ denotes spatial inversion and $1'$ time inversion. Differentiating the toroidal moment is that it is reversed by both spatial and time inversion. To determine the toroidal moment tensor invariant under the group **H** of a species **G1′***F***H**, we have tabulated the toroidal moment tensor invariant under each of the 122 magnetic point groups (see §3 of the supplementary material).[1] The coset representatives of the coset decomposition of **G1′** with respect to **H** were determined using software (Schlessman & Litvin, 2001). Using the form of the toroidal moment tensor invariant under **H** and the coset representatives of the coset decomposition of **G1′** with respect to **H**, we calculated the form of the toroidal moment in each of the domain states and subsequently determined the toroidal moment distinction of the domain states. A section of a new listing of the 773 species, each characterized by the distinguishability by all four primary ferroics, *i.e.* giving the distinction characterization by spontaneous toroidal moment, magnetization, polarization and strain, is given in Table 2, with the complete listing in the supplementary material.[1]

The first column gives the sequential numbering as found in Aizu (1970). The second column gives **G1′***F***H** with the groups given in a non-coordinate format with p and s denoting *principal* and *secondary* axes. In the third column, **G1′***F***H** is given in a format specifying the coordinate axes of **G** and **H**. The final four columns give, for each species, its *distinction quadruplet* and a set of four letters, representing the distinguishability of the domain states by a toroidal moment T, magnetization M, polarization P and strain ε_{ij}. Each physical property is assigned a letter representing its distinguishability type (Litvin, 1984):

F Full − distinguishes all n domain states

P Partial − distinguishes m, $1 < m < n/2$, domain states, *i.e.* some but not all domain states

N Null − does not distinguish any domain state and is non-zero

Z Zero − does not distinguish any domain state and is zero.

(1)

For example, the distinction quadruplet for species $4221'F41'$, see species 128 in Table 2, is Z Z F N. That is, spontaneous toroidal moment and magnetization vanish in the domain states, spontaneous polarization distinguishes the two domain states, and spontaneous strain is not zero and is the same in the two domain states.

In the fourth column, the number $n = |\mathbf{G1'}|/|\mathbf{H}|$ of domain states is given. This number is given in one of two formats depending on the group $\mathbf{H}$.

1. If $\mathbf{H} = \mathbf{K1'}$, *i.e.* the group $\mathbf{H}$ contains the time inversion $1'$, the number of single domain states is given simply as n. In such cases, the meaning of full, partial, null and zero follows the definitions in (1) above.

2. If the group $\mathbf{H}$ does not contain the time inversion $1'$, the number is given as $n/2 \times 2$. In these cases, the meaning of full and partial in (1) above for spontaneous polarization and spontaneous strain are replaced with

F Full − distinguishes $n/2$ domain states

P Partial − distinguishes m, $1 < m < n/2$, domain states,
   i.e. some but not all domain states.

$$(2)$$

This follows from the fact that time inversion can be taken as a coset representative and both polarizations and strain are invariant under time inversion. Consequently, for a group $\mathbf{H}$ not containing $1'$, a listing of 'full' under spontaneous polarization or spontaneous strain means there are not n but $n/2$ distinct domain states. For example, the species 114, $4221'F1$, with distinction quadruplet F F F F has the number of domain states listed as 8×2. For this species, spontaneous toroidal moment and magnetization distinguish all $n = 16$ domain

Figure 1
Distinction quadruplet classification of the 773 species of phase transitions. FT, PT and ZT denote full, partial and zero ferrotoroidics, FM, PM and ZM denote full, partial and zero ferromagnetics. The zero ferromagnetics are divided into two, the AFM antiferromagnetic case and the PDM para- and diamagnetic. FE, PE and NE denote full, partial and null ferroelastics. FP, PP and N/ZP denote full, partial and null or zero ferroelectrics. Each rectangular box represents a single ensemble. The pair of numbers $n(m)$ in the upper left corner of each rectangular represent the ensemble number n and the total number of species in that ensemble.

Table 3
The 21 species $\mathbf{G1'}F\mathbf{H}$ of phase transitions with distinction quadruplet Z Z F N.

The first six with $\mathbf{H}$ not containing time inversion $1'$ are AFM, antiferromagnetic cases, the remaining 15, with $\mathbf{H}$ containing time inversion, are PDM, para/diamagnetic cases.

Species No.	Species	
105	$4/m1'F4'$	$4_z/m_z1'F4'_z$
127	$4221'F4'$	$4_z2_z2_{xy}1'F4'_z$
240	$4/mmm1'F4'm'm$	$4_z/m_zm_xm_{xy}1'F4'_zm'_xm_{xy}$
354	$6/m1'F6'$	$6_z/m_z1'F6'_z$
381	$6221'F6'$	$6_z2_z2_11'F6'_z$
520	$6/mmm1'F6'm'm$	$6_z/m_zm_xm_11'F6'_zm'_xm_1$
3	$\bar{1}1'F1'$	$\bar{1}1'F1'$
21	$2/m1'F21'$	$2_z/m_z1'F2_z1'$
24	$2/m1'Fm1'$	$2_z/m_z1'Fm_z1'$
69	$mmm1'Fmm21'$	$m_xm_ym_z1'Fm_xm_y2_z1'$
106	$4/m1'F41'$	$4_z/m_z1'F4_z1'$
128	$4221'F41'$	$4_z2_x2_{xy}1'F4_z1'$
241	$4/mmm1'F4mm1'$	$4_z/m_zm_xm_{xy}1'F4_zm_xm_{xy}1'$
262	$\bar{3}1'F31'$	$\bar{3}_z1'F3_z1'$
271	$321'F31'$	$3_z2_x1'F3_z1'$
309	$\bar{3}m1'F3m1'$	$\bar{3}_zm_x1'F3_zm_x1'$
329	$\bar{6}1'F31'$	$\bar{6}_z1'F3_z1'$
355	$6/m1'F61'$	$6_z/m_z1'F6_z1'$
382	$6221'F61'$	$6_z2_z2_11'F6_z1'$
432	$\bar{6}m21'F3m1'$	$\bar{6}_zm_x2_11'F3_zm_x1'$
521	$6/mmm1'F6mm1'$	$6_z/m_zm_xm_11'F6_zm_xm_11'$

states. However, spontaneous polarization and strain distinguish only $n/2 = 8$ domain states.

If the toroidal moment distinguishability is removed from a distinction quadruplet, we have the distinction triplet used by Aizu (1970). For species No. 118, see Table 2, the distinction quadruplet is F F P F while the distinction triplet is F P F. As the distinction triplets were used by Schmid (1999) to classify the 773 species ensembles, we use the distinction quadruplets to classify the 773 species into 144 *sub-ensembles*: each sub-ensemble is characterized by a distinction quadruplet with the zero spontaneous magnetization distinguishability case divided into the two subtypes PDM and AFM. In Fig. 1, we show the 36 ensembles defined by Schmid and how each ensemble splits into sub-ensembles. This figure also gives the number of species in each ensemble and sub-ensemble. FT, PT and ZT denote full, partial and zero ferrotoroidics, FM, PM and ZM denote full, partial and zero ferromagnetics. The zero ferromagnetics are divided into two, the AFM antiferromagnetic case and the PDM para- and diamagnetic. FE, PE and NE denote full, partial and null ferroelastics. FP, PP and N/ZP denote full, partial and null or zero ferroelectrics. Each rectangular box represents a single ensemble. The pair of numbers $n(m)$ in the upper left corner of each rectangle represent the ensemble number n and the total number of species in that ensemble.

For example, there are 21 species of phase transitions with the distinction quadruplet Z Z F N, 6 AFM and 15 PDM cases. For each distinction quadruplet, we have also listed the corresponding species. As an example, in Table 3, we list the 21 species with the distinction quadruplet Z Z F N.[2] In Table 4,

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[2] See deposition footnote.

Table 4
Example of an ensemble, listing the associated sub-ensembles and sub-ensemble species.

Ensemble No.	Distinction triplet	No. of species
10	P F F	18

Sub-ensemble distinction quadruplet	No. of species	Sp. No.	Species	
F P F F	9	41	$mm21'Fm$	$m_x m_y 2_z 1'Fm_x$
		137	$4mm1'Fm$	$4_z m_x m_{xy} 1'Fm_x$
		158	$\bar{4}2m1'Fm$	$\bar{4}_z 2_x m_{xy} 1'Fm_{xy}$
		325	$\bar{6}1'Fm$	$\bar{6}_z 1'Fm_z$
		391	$6mm1'Fm$	$6_z m_x m_1 1'Fm_x$
		414	$\bar{6}m21'Fm(p)$	$\bar{6}_z m_x 2_1 1'Fm_z$
		415	$\bar{6}m21'Fm(s)$	$\bar{6}_z m_x 2_1 1'Fm_x$
		422	$\bar{6}m21'Fm'm2'(sp)$	$\bar{6}_z m_x 2_1 1'Fm_z m'_y 2'_1$
		617	$\bar{4}3m1'Fm$	$\bar{4}_z 3_{xyz} m_{xy} 1'Fm_{xy}$
P P F F	7	31	$2221'Fm$	$2_x 2_y 2_z 1'F2_z$
		83	$\bar{4}1'F2$	$\bar{4}_z 1'F2_z$
		117	$\bar{4}221'F2(s)$	$\bar{4}_z 2_x 2_{xy} 1'F2_x$
		153	$\bar{4}2m1'F2(s)$	$\bar{4}_z 2_x m_{xy} 1'F2_x$
		366	$6221'F2(s)$	$6_z 2_x 2_1 1'F2_x$
		535	$231'F2$	$2_x 3_{xyz} 1'F2_x$
		584	$4321'F2(s)$	$4_z 3_{xyz} 2_{xy} 1'F2_{xy}$
Z P F F	2	167	$\bar{4}2m1'Fm'm'2$	$\bar{4}_z 2_x m_{xy} 1'Fm'_{xy} m'_x 2_z$
		625	$\bar{4}3m1'Fm'm'2$	$\bar{4}_z 3_{xyz} m_{xy} 1'Fm'_{xy} m'_x 2_z$

we give an example of an ensemble, listing the associated sub-ensembles and sub-ensemble species, a complete listing for each ensemble is given in the supplementary material.[3]

APPENDIX A
Primary and secondary ferroic property tensors

Property tensors play a dual role when ferroic crystals are considered:

1. in characterizing the 773 species $\mathbf{G}1'F\mathbf{H}$;
2. in determining possible domain switching.

The species $\mathbf{G}1'F\mathbf{H}$ are characterized by the tensor form of physical properties of the species' associated domain states. The forms T_i, $i = 1, \ldots, n$, of a physical property tensor $\mathbf{T}$ in the domain states are determined, as explained above, by the group $\mathbf{H}$ and the coset representatives of the coset decomposition of $\mathbf{G}1'$ with respect to $\mathbf{H}$. Knowing these tensor forms then gives rise to the full, partial, null or zero characterization by physical properties of each species. All tensor forms are generated from the physical property's tensor form T_1, invariant under $\mathbf{H}$. We have tabulated in the supplementary material[3] the tensor form of the physical property toroidal moment, along with magnetization, polarization and strain, invariant under the 122 magnetic point groups (Opechowski, 1986).[3] [For the method used to determine the tensor forms invariant under the magnetic point groups from known

Table 5
Physical properties and corresponding free energy terms and ferroic types.

Physical property		Free-energy term	Ferroic type
			Primary ferroics
$\epsilon_{(s)ij}$	Spontaneous strain	$\Delta\varepsilon_{(s)ij}\sigma_{ij}$	Ferroelastic
$M_{(s)i}$	Spontaneous magnetization	$\Delta M_{(s)i}H_i$	Ferromagnetic
$P_{(s)i}$	Spontaneous polarization	$\Delta P_{(s)i}E_i$	Ferroelectric
$T_{(s)i}$	Spontaneous toroidal moment	$\Delta T_{(s)i}S_i$	Ferrotoroidic
			Secondary ferroics
s_{ijkl}	Elastic compliance	$\frac{1}{2}\Delta s_{ijkl}\sigma_{ij}\sigma_{kl}$	Ferrobielastic
χ_{ij}	Magnetic susceptibility	$\frac{1}{2}\Delta\chi_{ij}H_iH_j$	Ferrobimagnetic
κ_{ij}	Electric susceptibility	$\frac{1}{2}\Delta\kappa_{ij}E_iE_j$	Ferrobielectric
τ_{ij}	Toroidic susceptibility	$\frac{1}{2}\Delta\tau_{ij}S_iS_j$	Ferrobitoroidic
d_{ijk}	Piezoelectric coefficient	$\Delta d_{ijk}E_i\sigma_{jk}$	Ferroelastoelectric
q_{ijk}	Piezomagnetic coefficient	$\Delta q_{ijk}H_i\sigma_{jk}$	Ferromagnetoelastic
γ_{ijk}	Piezotoroidic coefficient	$\Delta\gamma_{ijk}S_i\sigma_{jk}$	Ferroelastotoroidic
α_{ij}	Magnetoelectric coefficient	$\Delta\alpha_{ij}H_iE_j$	Ferromagnetoelectric
ζ_{ij}	Magnetotoroidic coefficient	$\Delta\zeta_{ij}H_iS_j$	Ferromagnetotoroidic
θ_{ij}	Electrotoroidic coefficient	$\Delta\theta_{ij}E_iS_j$	Ferroelectrotoroidic

tabulations of the tensor forms invariant under non-magnetic point groups see Litvin (1994).]

In determining domain switching, one focuses on the differences of the forms of a physical property tensor $\mathbf{T}$ in a species' domain states (Schmid, 2003b; Newnham, 2005). The driving potential of domain switching is the difference ΔG in free energy of two domain states, which to second order in external fields is

$$
\begin{aligned}
\Delta G = {} & \Delta\varepsilon_{(s)ij}\sigma_{ij} + \Delta M_{(s)i}H_i + \Delta P_{(s)i}E_i + \Delta T_{(s)i}S_i \\
& + \tfrac{1}{2}\Delta s_{ijkl}\sigma_{ij}\sigma_{kl} + \tfrac{1}{2}\Delta\chi_{ij}H_iH_j + \tfrac{1}{2}\Delta\kappa_{ij}E_iE_j + \tfrac{1}{2}\Delta\tau_{ij}S_iS_j \\
& + \Delta d_{ijk}E_i\sigma_{jk} + \Delta q_{ijk}H_i\sigma_{jk} + \Delta\gamma_{ijk}S_i\sigma_{jk} + \Delta\alpha_{ij}H_iE_j \\
& + \Delta\zeta_{ij}H_iS_j + \Delta\theta_{ij}E_iS_j.
\end{aligned} \tag{3}
$$

$\Delta\varepsilon_{(s)ij}$, $\Delta M_{(s)i}$, $\Delta P_{(s)i}$, $\Delta T_{(s)i}$ are the difference of, respectively, the spontaneous strain, magnetization, polarization and toroidal moment in a pair of domain states. σ_{ij}, H_i, E_i and S_i are, respectively, the external stress, magnetic field, electric field and where $S_i \sim (E \times H)_i$ (Schmid, 2001). The coefficient ΔT of the combination of external fields in each term is the difference of a physical property $\mathbf{T}$ in a pair of domain states associated with a species $\mathbf{G}1'F\mathbf{H}$. If T_1 is the form of the physical property tensor invariant under $\mathbf{H}$, then $\Delta T = T_1 - g_jT_1$, where g_j is a coset representative of the coset decomposition of $\mathbf{G}1'$ with respect to $\mathbf{H}$, and is the difference of the physical property $\mathbf{T}$ in the pair of domain states S_1 and S_j. The coset representatives are given in Schlessman & Litvin (2001) and we have tabulated the form of all physical property tensors appearing in the above equation invariant under the 122 magnetic point groups.[4] The physical property tensors associated with the free-energy difference given above are listed in Table 5 with the term in which it appears in the equation of ΔG and the nomenclature of the type of ferroic

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[3] See deposition footnote.

[4] See deposition footnote.

whose domain states are distinguished by that physical property tensor.

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