mode at $q = 0$, with rotational eigenvector around [001]. This softening at $q = 0$ (which we observed in the X-ray diffuse scattering from our crystal) also causes an increase in total crystal entropy. We illustrate the free energy at room temperature in Fig. 3. The details of the figure are drawn quite arbitrarily. The $I\bar{4}3m$ free energy is drawn higher than $Fm3m$, to accord with experimental observation. The difference between the dotted and solid $F\bar{4}3m \rightarrow I\bar{4}3m$ curve is the notional entropy effect of the phonon softening.

Application to real adamantane crystals

(a) Solution or vapour grown crystals

An $Fm3m$ crystal is calculated to undergo a phase transition at 300 K to an ordered phase at a few percent positive tetragonal distortion. This phase may be best described as strained $F\bar{4}3m$, in which case there will be a second transition (with $\bar{\theta}_z$ changing from ~ 0 to $45°$) to strained $I\bar{4}3m$; or the transition may be direct to an $I\bar{4}3m$ phase. Only $Fm3m$ phases have been observed in unstrained systems (Lechner & Heidemann, 1976; Damien & Fouret, 1979; Windsor et al., 1978).

(b) Crystals grown by annealing

The above cannot explain the observation of $F\bar{4}3m$ in our annealed crystal (Reynolds, 1978) since we measure $T = 0 \pm \frac{1}{2}\%$. However, our crystal had a large measured mosaic spread ($6°$) about one axis only ([001]) due to the growth in a tube and was optically uniform.

We may surmise that polygonization of the crystal, initially saturated with dislocations along [001] (density $\sim 10^{15}$ m^{-2}), to give a block size of diameter ≤ 300 molecules, occurs. The maximum size is dictated by the optical uniformity of the crystal. Each block would be defined by an array of ≤ 60 dislocations around its

boundary. Such a small number would ensure substantial local tetragonal strain, of the order of 7% near the block boundary. This defect structure is compatible with our observations.

There are three possibilities for the structure of the crystal – all $Fm3m$, all $F\bar{4}3m$, or mixed $Fm3m$ (in regions of low T) and $F\bar{4}3m$ (in regions of high T). The pure phases will be stable if the correlation range of clusters of the other phase is at least comparable with the block size, if not, the two-phase system will be stable. Qualitatively, we can say that if the free-energy difference between the two phases is small, as it is, then the correlation range may be large enough to stabilize a single phase. The stable phase will be that which has the lowest free energy, given approximately by an averaging of the free-energy curves (Fig. 3) over the distribution of local tetragonal strain. While $Fm3m$ is favoured at $T \leq 0$, it is strongly disfavoured at high T. Therefore, a crystal including regions of large positive tetragonal strain may well adopt an $F\bar{4}3m$ phase. A more definite prediction requires far more knowledge of the crystal's imperfections and the thermodynamics of the pure phases.

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Gyrotropy: An Implicit Form of Ferroicity

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Abstract

Some optically active ferroic crystals (a few ferroelectrics and one ferroelastic) are known to exhibit reversal of the sign of spontaneous optical rotation in certain directions when a suitable electric field or uniaxial stress is applied. This phenomenon, to be called gyrotropy, is examined in the framework of Aizu's

formalism of ferroicity. The case of dicalcium strontium propionate, $Ca_2Sr(C_3H_5O_2)_6$, a ferroelastic–ferroelectric, is analysed to illustrate a simple procedure for enumerating gyrotropic orientation states. The 18 'pure' gyrotropic species of crystals are listed: these are those species for which gyrotropy is not accompanied by primary ferroicity. Ni–Cl boracite, $BaMnF_4$, $RbNO_3$, and some related crystals, are suggested as new potential gyrotropics. The first two of these are ferromagnetic gyrotropics, of which no examples are known so far.

1. Introduction

A crystal is said to be ferroic if it is capable of possessing domain structure and if the domain walls can be moved (at least in some limited temperature range) by the application of a suitable driving force such as electric field, magnetic field, uniaxial stress, or any combination of these. The ferroic phase results through a small lattice distortion from a supergroup phase (called the prototype), with the accompanying loss of some symmetry elements at the phase transition. It therefore has two or more stable orientation states (domains), which can be mapped onto one another by those symmetry operations which disappear at the phase transition (Janovec & Shuvalov, 1975; Wondratschek & Jeitschko, 1976).

The concept of ferroicity was developed in a unified and generalized way by Aizu (1970a), after he had added the new and powerful idea of ferroelasticity to the two earlier, well-known types of (primary) ferroicity, namely ferroelectricity and ferromagnetism. Aizu derived and tabulated 773 possible ferroic species (Aizu, 1970a; see also Cracknell, 1972). By examining the effect of the additional symmetry elements present in the prototype point group on the symmetry-adapted property tensors of the ferroic phase, different group–subgroup pairs (species) were labelled as either full, or partial, or non-ferroelectric, -ferromagnetic, and/or -ferroelastic. Secondary (and higher-order) ferroicity, which can be important mainly when the corresponding primary ferroicity is absent, was later discussed by Aizu (1973), and also by Newnham & Cross (1974a,b).

Spontaneous polarization, spontaneous magnetization, and spontaneous strain present in primary ferroics are, respectively, first-rank-polar, first-rank-axial, and second-rank-polar tensor properties. It is possible, however, that at a phase transition a spontaneous second-rank-axial tensor property also develops non-zero components, and if its representation surface can be re-oriented (switched) to equivalent configurations by suitable driving forces, we would have a fourth kind of 'primary' ferroicity. A property of this type is the optical gyration tensor, (g_{ij}) (Nye, 1964; Ramachandran & Ramaseshan, 1961).[*]

Application of a driving force to a ferroic crystal makes some domains more stable than others. State shifts between two domains occur when the driving force is such that the difference in the stored free enthalpy in the two domains is large enough to overcome the energy barrier between them. Optical activity, however, arises from a wavevector-dependent term in the expression for the general dielectric tensor, whose contribution to the stored enthalpy is zero (Juretschke, 1974). Therefore, changes of free enthalpy during gyrotropic state shifts get their entire contribution from the concomitant electric, magnetic and/or mechanical ferroic state shifts of first or higher order. For this reason, gyrotropy, as mentioned above in the context of optical gyration, should be regarded as only an implicit form of ferroicity. Gyrotropic state shifts can occur only in those crystals which are ferroic (in the explicit sense). We therefore suggest the following definition: a gyrotropic crystal may be defined as a *ferroic* (primary or of higher order), of which at least two orientation states have gyration tensors differing in the signs (or at least the magnitudes) of one or more of their corresponding components.

Although the above definition and also the discussion to follow are in terms of the gyration tensor, one could define a gyrotropic state shift in a more general way as one in which the representation surface of *any* second-rank-axial tensor property flips to an equivalent orientation. Apart from the gyration tensor, another important axial second-rank property tensor is the linear magnetoelectric susceptibility tensor (Freeman & Schmid, 1975). However, whereas the gyration tensor is invariant under a time-inversion operation, the magnetoelectric tensor is not. The magnetoelectric effect occurs only in some magnetic (or time-asymmetric) crystals and, unlike optical activity, magnetoelectricity is not an implicit but an explicit form of (secondary) ferroicity. Thus, to maintain uniformity, it is advisable to define gyrotropy, as we have done, only with reference to optical activity, even for crystals which can show the magnetoelectric effect.

2. Gyrotropic crystals

Gyrotropic crystals can be conveniently categorized according to their concomitant (explicit) ferroic property.

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[*] In the original version of the present paper, two alternative names, 'ferroenantiomorphism' and 'ferrogyration' were suggested for the phenomenon where the sign of spontaneous optical rotatory power of a crystal can be reversed in sign in certain directions by applying suitable driving forces. But the author was then unaware of a very recent paper, *Gyrotropic Phase Transitions* by Koňák, Kopský & Smutný (1978) on the same general topic. To avoid confusion in the literature, these names have therefore been withdrawn in favour of the term 'gyrotropy'.

(i) Ferroelastics

Dicalcium strontium propionate (DSP), $Ca_2Sr(C_2H_5COO)_6$, is the only reported case of a ferroelastic crystal exhibiting reversal of optical rotation under the application of appropriate uniaxial stress. It is also a ferroelectric below 281·7 K. The ferroelastic behaviour of DSP was first observed and explained by Sawada, Ishibashi & Takagi (1977), who assigned this crystal to the Aizu species $m3m\, F\, 422$. [In this notation (Aizu, 1970a), F stands for 'ferroic', and the symbols on its left and right are, respectively, the point-group symmetries of the prototype and ferroic phases; we omit $1'$, denoting time-inversion symmetry, from the original Aizu symbol when discussing non-magnetic crystals.] The group–subgroup relationship between the prototype and ferroic phases of DSP can be expressed by the following direct product:

$$(m3m) = (422) \times (3) \times (\bar{1}). \qquad (1)$$

$(m3m)$ and (422) are groups of order 48 and 8 respectively, so that 48/8 or six orientation states are possible (Aizu, 1970a). However, there are only three distinct ferroelastic orientation states [$S_1(\sigma)$, $S_2(\sigma)$, $S_3(\sigma)$, say], corresponding to the three symmetry elements of the group (3). This is because the strain tensor, (σ_{ij}), is invariant under operations of the inversion group $(\bar{1})$. For the gyration tensor, (g_{ij}), on the other hand, all six states are distinct. Three of these: $S_1(g)$, $S_2(g)$, $S_3(g)$, are the same as $S_1(\sigma)$, $S_2(\sigma)$, $S_3(\sigma)$, respectively, because the groups (422) and (3) comprise only proper rotations, and under the proper rotations of (3), tensors (σ) and (g) transform in the same way. They may be explicitly written as follows (Nye, 1964):

$$S_1(g) = \text{diag}\, (b\, b\, c), \qquad (2)$$

$$S_2(g) = \text{diag}\, (c\, b\, b), \qquad (3)$$

$$S_3(g) = \text{diag}\, (b\, c\, b). \qquad (4)$$

Here $b = g_{11} = g_{22}$ and $c = g_{33}$, and diag $(b\, b\, c)$ denotes a diagonal matrix with b, b, c as the diagonal elements. The three additional gyrotropic states, $S_4(g)$, $S_5(g)$, $S_6(g)$, are obtained by applying the inversion operation to the states S_1, S_2, S_3, respectively. We get

$$S_4(g) = \text{diag}\, (-b\, -b\, -c), \qquad (5)$$

$$S_5(g) = \text{diag}\, (-c\, -b\, -b), \qquad (6)$$

$$S_6(g) = \text{diag}\, (-b\, -c\, -b). \qquad (7)$$

It should be possible to effect ferroelastic state shifts among S_1, S_2, S_3, and gyrotropic state shifts between any one of S_1, S_2, S_3 and any one of S_4, S_5, S_6, by applying the required uniaxial stress. State shifts in which no gyration tensor component changes sign [like $S_1(g) \leftrightarrow S_2(g)$, or $S_4(g) \leftrightarrow S_6(g)$] are indistinguishable from the corresponding ferroelastic state shifts, and should be referred to as ferroelastic state shifts only.

Sawada, Ishibashi & Takagi (1977) have observed state shifts in DSP in which the tetragonal c axis is rotated by 90° and, simultaneously, the sense of optical rotation is reversed. This corresponds, in the above notation, to the state shifts $S_1(g) \leftrightarrow S_6(g)$. It is interesting to note that purely ferroelastic state shifts of the type $S_1(g) \leftrightarrow S_3(g)$, in which there is no reversal of the sign of optical rotation in any direction, are never observed, although from symmetry considerations they are possible.

Let us now consider the ferroelectric phase of DSP. The ferroelectric phase transition at 281·7 K lowers the symmetry from 422 to 4 (Kobayashi & Yamada, 1962). However, contrary to the statement by Aizu (1973) (which was made before the ferroelastic effect in this crystal was demonstrated), the prototype symmetry cannot be chosen as 422. This is because a prototype should not be derivable from a still higher symmetry phase (real or hypothetical) by a small lattice distortion (Aizu, 1978), whereas Sawada, Ishibashi & Takagi (1977) found it expedient to postulate $m3m$ as the prototype symmetry to explain the ferroelastic behaviour of this crystal. The ferroelectric phase of DSP should therefore be assigned to the species $m3m\, F\, 4$, and not $422\, F\, 4$.

For this species, 48/4 or 12 orientation states are possible. It is a partial-ferroelectric partial-ferroelastic (Aizu, 1970a). It is also a *partial gyrotropic*, because the number of distinct gyration tensor states is still six, the same as in the species $m3m\, F\, 422$ above 281·7 K. (This can be easily deduced by using equation 8 below.)

An interesting observation made by Kobayashi & Yamada (1962) was the occasional 'spontaneous' racemization that occurs in DSP in the ferroelectric phase. The resultant *dextro* $\leftrightarrow$ *laevo* transformation of domains, with the tetragonal c axes in the two types of domains remaining parallel, is easily understood in terms of symmetry operations. We have:

$$(m3m) = (4) \times (3) \times (2/m), \qquad (8)$$

where (4) is the symmetry of the ferroelectric phase. The gyrotropic $d \leftrightarrow l$ state shifts (racemization), in which the c axes remain parallel, are accounted for by the symmetry operation m of the group $(2/m)$. Incidentally, this is a fact which cannot be explained by choosing 422 as the prototype symmetry.

Ferroelastic domain walls generally have the possibility of occurring as mutually perpendicular pairs of planar surfaces (Sapriel, 1975). This is a consequence of strain compatibility requirements between contiguous domains and of the fact that spontaneous strain tensors can be expressed as traceless matrices (Aizu, 1970b). For the ferroelectric phase of DSP there is no strain compatibility requirement to be fulfilled for the above mentioned enantiomorphous domains related by the symmetry operation m, because the two domains

have identical spontaneous strain tensors. The polarization vectors in the two domains are also parallel, so that no strains at the domain walls arise from this cause also. Consequently, these domain walls are not expected to be either planar or perpendicular in pairs (Janovec & Shuvalov, 1975). This is indeed found to be the case. The domain walls photographed by Kobayashi & Yamada (1962) are completely irregular in shape and orientation. This is also the case in the ferroelectric gyrotropic $Pb_5Ge_3O_{11}$ (Newnham & Cross, 1974c). In $CuCsCl_3$ (Hirotsu, 1975) also, a ferroelastoelectric crystal, wedge-shaped gyrotropic domain boundaries are observed. Interesting inversion domains with irregularly oriented boundaries have also been observed in the χ-phase intermetallic compound $Fe_{36}Cr_{22-x}(Ti,Mo)_x$, and several other compounds like γ-brass, δ-NiMo, lithium ferrite, and Al_4Mo (Amelinckx, 1976).

Regarding new ferroelastic gyrotropics, we may mention the trigonal polymorphs of $RbNO_3$, $CsNO_3$ and $TlNO_3$ as likely candidates, although ferroelasticity has not yet been demonstrated in these crystals (see Bury & McLaren, 1969, and the references therein). These crystals belong to the species $m3m\,F\,3$, which is a partial-ferroelectric partial-ferroelastic species, meaning that primary as well as higher-order state shifts are possible (Aizu, 1972).

(ii) Ferroelectrics

A good discussion of known ferroelectric gyrotropics is given in the recent paper of Koňák, Kopský & Smutný (1978).

(iii) Ferromagnetics

No optically active magnetically ordered materials appear to have been investigated to observe any reversal of spontaneous optical rotation when magnetic state shifts occur. A class of crystals in which this effect can be expected to occur is represented by Ni–Cl boracite, $Ni_3B_7O_{13}Cl$. Below 9 K, this transparent crystal belongs to the ferroic species $\bar{4}3m1'\,F\,m'm2'$ (Rivera, Schmid, Moret & Bill, 1975; Schmid, 1975), and is therefore a full-ferromagnetic, full-ferroelectric, full-ferroelastic, with 12 orientation states (Aizu, 1970a). Similarly, Ni–I boracite is also a potential gyrotropic. It is a pale green transparent crystal, exhibiting weak ferromagnetism and magneto-electricity below 64 K. Another possible crystal is $BaMnF_4$, which is also transparent (pink) and shows the magnetoelectric effect (Scott, 1978). Below 25 K it belongs to the species $mmm1'\,F\,2$ (Abrahams & Keve, 1971), which is a partial-ferromagnetic, partial-ferroelectric, partial-ferroelastic species.

(iv) Secondary and higher-order ferroics

Koňák, Kopský & Smutný (1978) have suggested some crystals in which 'pure' gyrotropic phase transitions can be expected, that is, those accompanied by the onset of neither ferroelectricity, nor ferroelasticity (nor ferromagnetism). Such crystals can undergo only second or higher orders of electrical, mechanical, or electromechanical state shifts (Aizu, 1972), and will, therefore, generally require higher driving forces for the motion of the enantiomorphous domain walls than are necessary for primary ferroics (Newnham & Cross, 1974b). None of the possible pure gyrotropics listed by Koňák, Kopský & Smutný (1978) will allow electrical state shifts of order less than three, and mechanical state shifts of order less than two. All of them, except one, are ferroelastoelectrics (electromechanical order of state shifts = 2). The exception is ZrOS, belonging to the species $\bar{4}3m\,F\,23$, for which the electrical order of state shifts is five or higher, mechanical order of state shifts is three or higher, and the electromechanical order is three or higher.

Koňák, Kopský & Smutný (1978) have also derived and listed in Table 1 of their paper the possible species of pure gyrotropics resulting from non-gyrotropic prototype phases. The same list can also be obtained by inspection from Table 1 of Aizu (1970a). But such an inspection shows that Table 1 of Koňák, Kopský & Smutný (1978) is incomplete. There should be five additional species of pure gyrotropics: $\bar{4}2m\,F\,\bar{4}$, $4/mmm\,F\,\bar{4}$, $6mm\,F\,3$, $6/mmm\,F\,32$, and $m3m\,F\,23$. Of these, $\bar{4}2m\,F\,\bar{4}$ has a gyrotropic prototype and so falls outside the scope of Table 1 of Koňák, Kopský & Smutný (1978), but the remaining four should find a place in it. We give here, for the convenience of the reader, a complete list of 18 possible pure-gyrotropic, non-magnetic, ferroic species. All of them except one (No. 4) have non-gyrotropic prototypes.

1.	$mmm\,F\,222$	10.	$6mm\,F\,3$
2.	$4/m\,F\,\bar{4}$	11.	$6mm\,F\,6$
3.	$4mm\,F\,4$	12.	$\bar{6}m2\,F\,32$
4.	$\bar{4}2m\,F\,\bar{4}$	13.	$6/mmm\,F\,32$
5.	$4/mmm\,F\,\bar{4}$	14.	$6/mmm\,F\,622$
6.	$4/mmm\,F\,422$	15.	$m3\,F\,23$
7.	$4/mmm\,F\,\bar{4}2m$	16.	$\bar{4}3m\,F\,23$
8.	$3m\,F\,3$	17.	$m3m\,F\,23$
9.	$\bar{3}m\,F\,32$	18.	$m3m\,F\,432$

Species No. 7 is split by Koňák, Kopský & Smutný (1978) into two: $4/mmm\,F\,\bar{4}_z2_xm_{xy}$ and $4/mmm\,F\,\bar{4}_zm_x2_{xy}$.

The four species left out by Koňák, Kopský & Smutný (1978) have four orientation states each, whereas the remaining 14 have only two each. These four species are the partial gyrotropics in the class of pure gyrotropics. We have already come across in DSP an example of a partial gyrotropic in the class of ferroelectric–ferroelastic gyrotropics.

3. Applications

Applications of ferroelectrics and ferromagnetics are well-known, and those of ferroelastics have been attempted (see, for example, Kumada, 1973 and the references therein). The additional degree of freedom offered by the reorientability of the gyration surface, particularly in primary ferroics, and the continuous variability of optical rotation with crystal thickness, optical wavelength, and poling, should be exploitable to advantage. Chern & Phillips (1972) have examined the possibility of using $NaNO_2$ ferroelectric gyrotropic crystals in information-storage devices, using the gyrotropic property. The use of the same property in $Pb_5Ge_3O_{11}$, another ferroelectric gyrotropic, for optical storage and optical signal processing has been discussed by Newnham & Cross (1974c), who also point out that since frequency dispersion is not strong for optical rotation, the use of optical rotation domains is preferable to birefringence domains for information storage devices, because then one can use white light in place of monochromatic light.

Apart from their possible practical applications, gyrotropic materials are also of great basic interest because optical activity studies can often be an additional way of monitoring their phase transitions.

Note added in proof. In a personal communication Dr Kopský points out that table 1 of Koňák, Kopský & Smutný (1978) gives only those pure gyrotropic species which involve equitranslational phase transitions associated with a single transition parameter. Thus the scope of their Table 1 is more limited than ours. The 18 species listed in the present paper exhaust *all* possible pure gyrotropic non-magnetic species.

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