OPTICAL PROPERTIES OF MAGNETIC CRYSTALS

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Received April 7, 1971

Abstract

From symmetry considerations and using generalized Onsager relations, it is shown that 66 of the 90 magnetic classes, consisting of 29 single colour and 37 double colour ones, can exhibit what may be called the strain gyrotropic rotation. Similarly, 69 of the 90 magnetic classes, consisting of 21 single colour and 48 double colour ones, can exhibit what may be called the strain gyrotropic birefringence. A crystal in the class m3 or m3$\underline{m}$ is interesting as it can exhibit strain gyrotropic rotation despite its being cubic and incapable of exhibiting gyrotropic rotation in the unstressed state. Similarly, a crystal in the class $\underline{m}$3$\underline{m}$, is interesting as it can exhibit strain gyrotropic birefringence despite its being cubic and incapable of exhibiting gyrotropic birefringence in the unstressed state.

1. Introduction

The induction D_i may be written in the general case as in (1), consisting of two terms, the first involving the components E_i of the incident electric field and the second involving the derivatives thereof.

$$D_i = a_{ik}E_k + \gamma_{ikl} \frac{\partial E_k}{\partial x_l} . \tag{1}$$

The usual summation convention is implied and all suffixes take values 1, 2 and 3. In earlier concepts, when no note was taken of the existence of magnetic point groups, the intrinsic symmetry of a_{ik} (dielectric tensor) and γ_{ikl} (gyration tensor) was assumed to be $a_{ik} = a_{ki}$ and $\gamma_{ikl} = -\gamma_{kil}$. D and E are respectively the electric induction and electric field vectors.

One can show that in nonmagnetic crystals, only the symmetric part of a_{ik} (*i.e.*, a^s_{ik}) and only the antisymmetric part of γ_{ikl} (*i.e.*, γ^a_{ikl}) survive because these crystals have time reversal as a symmetry operation and belong to what may be called the 'grey classes'. Thus, the earlier assumptions relating to intrinsic symmetry do cover all cases other than magnetic point groups, *i.e.*, the 'single colour and double colour classes'. In the latter, time reversal is not a symmetry operation and $\alpha^{a}{}_{ik}$ and $\gamma^{s}{}_{ikl}$ do not necessarily vanish. $\alpha^{a}{}_{ik}$ causes a rotation of the plane of polarisation, which by analogy has been called the gyrotropic rotation. Such rotation, unlike natural optical rotation, does not depend on the wave normal direction. $\gamma^{s}{}_{ikl}$ causes a birefringence, which by analogy has been called the gyrotropic birefringence. Such birefringence, unlike natural birefringence, depends on the wave normal direction. In an earlier paper by the author (1969), using generalized Onsager relations, it was shown that $\alpha^{a}{}_{ik}$ can be non-zero in 31 and that $\gamma^{s}{}_{ikl}$ can be non-zero in 66 of the magnetic classes. It was also concluded that $\bar{4}\,3\,\mathrm{m}$ and $\underline{\mathrm{m}}\,3\,\mathrm{m}$ are two interesting cubic magnetic classes in which, while natural birefringence, gyrotropic rotation and optical rotation vanish, gyrotropic birefringence ($\gamma^{s}{}_{ikl}$) does not. Thus, if one can find a suitable transparent crystal of either class and work with it in its magnetic phase, one may expect it to exhibit birefringence of this special kind. The class $\underline{\mathrm{m}}\,3$ is also exactly on the same footing but was not cited in that paper.

Since publishing the paper just referred to, the author has been examining some other optical properties, such as strain optic coefficients (Photoelasticity) and strain optic rotation (effect of strain on optical rotation) and studying how properties analogous to these are likely to show up in magnetic classes of crystals.

It is clear that while the effect of strain on ordinary birefringence has been called photo-elasticity and described by strain optic coefficients, we should expect to find also the analogous effect of strain on gyrotropic birefringence in magnetic classes and we may designate it as strain gyrotropic birefringence. Similarly, the effect in magnetic classes analogous to strain optic rotation in nonmagnetic classes will be called the strain gyrotropic rotation. Some interesting results that may be expected in regard to these new properties are reported in this paper.

2. METHOD

In several earlier publications by the author (1966), it has been shown that in respect of any physical property, the number of surviving constants for a crystal class can readily be evaluated, if the rank of the tensor appropriate to the physical property, its intrinsic symmetry, its type, *i.e.*, axial or polar and so on are all known. A number of physical properties, both magnetic and non-magnetic, have already been studied by such methods and the results published. In Table I are given the relevant features for eight different physical property tensors, specially chosen for being cited in this paper because they relate to optical properties leading to new results in magnetic classes of crystals. As in the earlier work, character expressions are obtained on the basis of generalized Onsager relations. α^{s}_{ik} and γ^{a}_{ikl} are quite common and the results in respect of them are well known. α^{a}_{ik} and γ^{s}_{ikl} have been discussed recently by the author (1969). p_{ijkl} is the photo-elasticity tensor and was the subject of detailed study by the author many years ago. g_{ijkl} has been studied by Ramaseshan and Ranganath (1969) in a recent paper. G_{ikl} and P_{ijklm} are new and represent the effect of strain on gyrotropic rotation and gyrotropic birefringence respectively. The results for these two properties are given here for the first time.

TABLE I

Property	Symbol	Type	Character X (R)
Natural birefringence	α^{s}_{ik}	Ordinary	$(4\cos^2\phi \pm 2\cos\phi)$
Gyrotropic rotation	α^{a}_{ik}	Magnetic	$\pm(\pm 2\cos\phi + 1)$
Optical rotation	γ^{a}_{ikl}	Ordinary	$(\pm 4\cos^2\phi + 2\cos\phi)$
Gyrotropic birefringence	γ^{s}_{ikl}	Magnetic	$\pm(8\cos^3\phi \pm 8\cos^2\phi + 2\cos\phi)$
Strain optic coefficients or photo-elasticity	p_{ijkl}	Ordinary	$(16\cos^4\phi \pm 16\cos^3\phi + 4\cos^2\phi)$
Strain optical rotation	g_{ijkl}	Ordinary	$(\pm 16\cos^4\phi + 16\cos^3\phi \pm 4\cos^2\phi)$
Strain gyrotropic rotation	G_{ikl}	Magnetic	$\pm(\pm 8\cos^3\phi + 8\cos^2\phi \pm 2\cos\phi)$
Strain gyrotropic birefringence	P_{ijklm}	Magnetic	$\pm(32\cos^5\phi \pm 48\cos^4\phi + 24\cos^3\phi \pm 4\cos^2\phi)$

In Table I, it may be noted that for all four magnetic type tensors, the alternative ± appears outside the bracket in additon to the alternative ± appearing inside the bracket. In the former case, the + and – are adopted respectively according as R is an operation or an anti-operation and in the latter, the

+ and − are adopted respectively according as R is a pure rotation operation or a rotation-reflection operation. These conventions have been explained in earlier publications. Detailed numbers of non-vanishing components are calculated for all the 90 magnetic classes but are not given here. In Table II are summarised the principal results in respect of the four optical properties which become significant in magnetic classes. In Table III are given the actual numbers for the eleven cubic magnetic classes, because these are of interest from an experimental point of view.

TABLE II

Symbol	Property	Classes in which the effect is permissible
$\alpha^a{}_{ik}$	Gyrotropic rotation	13 single colour (pyromagnetic) classes and 18 double colour (pyromagnetic) classes. There is no cubic class amongst these.
$\gamma^s{}_{ikl}$	Gyrotropic birefringence	20 single colour classes and 46 double colour classes. There are 5 cubic classes amongst these.
G_{ikl}	Strain gyrotropic rotation	29 single colour classes and 37 double colour classes. There are 5 cubic classes amongst these.
P_{ijklm}	Strain gyrotropic birefringence	21 single colour classes and 48 double colour classes. There are 8 cubic classes amongst these.

3. SOME CONCLUSIONS

Ramaseshan and Ranganath (1969) have already reported the somewhat unexpected result that the classes 4 mm, 3 m, $\bar{6}$, $\bar{6}$ m2, 6 mm and $\bar{4}$3 m which do not show optical activity have surviving coefficients in respect of the strain optical rotation tensor. The physical significance is that stress induces optical activity in these classes which are not originally optically active. Amongst these, $\bar{4}$3 m belongs to the cubic system and may be regarded as specially significant. Analogous to this is the result reported in the present work that two of the cubic magnetic classes, m3 and m3$\underline{\mathrm{m}}$, which do not show gyrotropic rotation ($\alpha^a{}_{ik}$) have one surviving coefficient each in respect of the strain gyrotropic rotation tensor (G_{ikl}),

TABLE III

Physical property/ Crystal class	α^{a}_{ik}	γ^{s}_{ikl}	G_{ikl}	P_{ijklm}
23	0	1	1	8
m3	0	0	1	0
$\underline{m}3$	0	1	0	8
$\bar{4}3m$	0	1	0	5
$\underline{\bar{4}}3\underline{m}$	0	0	1	3
432	0	0	0	3
$\underline{4}3\underline{2}$	0	1	1	5
m3m	0	0	0	0
$\underline{m}3\underline{m}$	0	0	0	3
$m3\underline{m}$	0	0	1	0
$\underline{m}3m$	0	1	0	5

The physical significance is that stress induces gyrotropic rotation in these classes which are not originally capable of exhibiting gyrotropic rotation. There are other magnetic classes of this type but m3 and $m3\underline{m}$ are specially singled out for mention because they belong to the cubic system and besides gyrotropic rotation being zero, the symmetry is such that neither gyrotropic birefringence (γ^{s}_{ikl}) nor strain gyrotropic birefringence (P_{ijklm}) can exist in them as may be seen from the results given in Table III. This means that if one can find a suitable transparent crystal of either class, namely m3 and $m3\underline{m}$, and work with it in its magnetic phase, one may expect it to exhibit gyrotropic rotation when stress is applied while in the absence of stress the crystal should be free from ordinary optical activity as well as gyrotropic rotation. Further, experimental observation of such a phenomenon, if exists, will not be complicated by any type of birefringence because, in these two classes, birefringence—ordinary or gyrotropic—cannot appear with or without a stress.

In a similar manner, the magnetic cubic class $\underline{m}\ 3\ \underline{m}$ is of special interest as it may be seen from Table III that in this, all tensors except P_{ijklm} vanish. This means that any type of optical activity—ordinary or gyrotropic—cannot exist in this class nor can it appear on the application of a stress. Consequently, if one can find a suitable transparent crystal of the class $\underline{m}\ 3\ \underline{m}$ and work with it in its magnetic phase, one may expect it to exhibit gyrotropic birefringence when stress is applied while in the absence of stress, the crystal should be free from ordinary as well as gyrotropic birefringence. We may note that the experimental observation of such a phenomenon, if it exists, will not be complicated by the simultaneous appearance of any kind of optical rotation.

REFERENCES

Bhagavantam, S. .. *Materials Res. Bull.*, 1969, **4**, 477.

———— *Crystal Symmetry and Physical Properties*, 1966, Academic Press London.

Ramaseshan, S. and Ranganath, G. S. *Physics of the Solid State*, 1969, Academic Press, London.