FARADAY ROTATION TENSOR-EXTRACTION OF COMPONENTS

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Abstract

This paper reports methods of obtaining the components of Faraday rotation tensor in anisotropic crystals, using longitudinal and transverse Faraday rotation along the optic axis. It is found that in crystals belonging to triclinic, monoclinic and orthorhombic symmetry, one can get 6 out of 9, 3 or 4 out of 5, and 2 out of 3 components respectively. In the case of uniaxial crystals only 1 component can be obtained. By measuring Faraday rotation in a randomly oriented polycrystal one more component in all the above classes can be obtained.

1. Introduction

Recently, Legall and Jamet (1971) have shown that the different phenomena associated with magneto-optics of magnetic crystals like Faraday, Kerr, Cotton-Mouton and Spin-Raman effects are not processes distinctly different from each other but really various manifestations of the spin-photon interaction. Faraday effect which is usually viewed as arising from circular birefringence, can be described as a quantum process involving an elastic scattering of photons by the magnetic spins with a $\pi/2$ spatial rotation in the polarization vectors of photons, while the Spin-Raman effect arises from an inelastic scattering and the two phenomena are very intimately related. In the case of cubic crystals exact analytical relations between these have also been obtained by these authors. It would therefore be of some interest to obtain the components of Faraday-rotation tensor in crystals of lower symmetry. This paper deals with the problem of extracting these components.

When a transparent isotropic medium is placed in a magnetic field, it rotates the plane of polarization of light traversing it along the lines of force.

This magnetic optic rotation or Faraday effect differs from natural optical activity in that the sense of rotation depends not only on the direction of light propagation but also on the direction of the magnetic field. The rotation is given by

$$\rho = VHL$$

where H is the magnetic field, L the length of the specimen and V the Verdet constant which represents rotation per unit length per unit magnetic field. [For a review on Faraday rotation in diamagnetic crystals *see* Ramaseshan and Sivaramakrishnan (1958) and Ramachandran and Ramaseshan (1961).]

In an anisotropic medium, however, the Verdet constant, V, changes with direction. Voigt (1908) considered this problem from a simple electron theory and the concept of anisotropic polarizability and showed that in certain types of monoclinic crystals in which the optic axes lie on the plane of symmetry, the magneto-optic rotation along the two optic axes may be different for the same applied field. Voigt himself demonstrated this beautifully (1908) in the case of cane sugar. He also foresaw the possibility of Verdet constant varying with direction in paramagnetic anisotropic crystals. This effect was experimentally shown by Becquerel [(1908), (1929)] who by an ingeneous experiment observed variation of Verdet constant with direction in anisotropic paramagnetic crystals.

The problem of Faraday rotation in anisotropic crystals was considered in detail theoretically by Le Corre (1957). He showed Faraday rotation to be representable by an asymmetric second rank polar tensor having 9 independent components for the triclinic system. He also worked out the forms of the Faraday rotation matrices. However, the actual methods of extracting the tensor components were not considered. This problem is made difficult because the magneto-optic rotation in crystals is usually measured along the optic axis. Although attempts have been made to measure rotations in directions away from the optic axis [Chauvin (1886) (for Calcite upto 3°) and by Ramaseshan (1951) (for Alumina upto 10°)] the strong linear birefringence affects the measurements. This therefore restricts the number of components that can be extracted. However, in crystals of orthorhombic and lower symmetry we also have another interesting effect, namely, the transverse Faraday rotation wherein one can observe rotation normal to the direction of the magnetic field. This effect together with the familiar longitudinal Faraday rotation helps one in extracting a number of tensocomponents in anisotropic crystals. In this paper methods of extracting the various Faraday rotation tensor components have been reported.

2. Faraday Rotation in Crystals

Le Corre (1957) has published the theory of Faraday rotation in anisotropic crystals. The theory is presented here briefly but in a slightly different form. In the presence of a magnetic field **H**, the induction **D** and the external field **E** of the light wave are related by the following equation:

$$\mathbf{D} = (\epsilon)\,\mathbf{E} + i\,(\rho)\,\mathbf{E}. \tag{1}$$

If the medium is transparent (ϵ) is a symmetric second rank tensor with real components and (ρ) is an antisymmetric tensor of second rank again with real components. If the medium is optically active, part of the rotation results from natural optical activity. In such optically active crystals by measuring rotation in the presence and the absence of **H**, rotation due to magnetic field **H** alone can be separated out. Hence for purely magneto-optic rotation

$$\mathbf{D} = (\epsilon)\,\mathbf{E} + i\,(\rho^f)\,\mathbf{E}. \tag{2}$$

The antisymmetric tensor (ρ^f) exists only when **H** is present. This can be replaced by a vector operator $\mathbf{G}^f \times$. Hence

$$\mathbf{D} = (\epsilon)\,\mathbf{E} + i\mathbf{G}^f \times \mathbf{E} \tag{3}$$

Alternatively if **E** is expressed as a function of **D** we get

$$\mathbf{E} = (a)\,\mathbf{D} - i\boldsymbol{\Gamma}^f \times \mathbf{D} \tag{4}$$

where Γ^f is called the magneto-gyration vector which is a function of **H** only. In a first order theory we can take

$$\boldsymbol{\Gamma}^f = (\Gamma^f)\,\mathbf{H} \tag{5}$$

where (Γ^f) is a general nine-component tensor.

If we solve Maxwell's equations for such a medium, we find that along any direction s two crossed elliptic vibrations travel with velocities v' and v'' given by

$$\left.\begin{aligned} v'^2 &= \tfrac{1}{2}(v_1^2 + v_2^2) - \tfrac{1}{2}\left|\sqrt{(v_1^2 - v_2^2)^2 + 4\gamma^2}\right| \\ v''^2 &= \tfrac{1}{2}(v_1^2 + v_2^2) + \tfrac{1}{2}\left|\sqrt{(v_1^2 - v_2^2)^2 + 4\gamma^2}\right| \end{aligned}\right\} \tag{6}$$

where v_1 and v_2 are the principal velocities in the absence of the field **H** and $\gamma = \Gamma_{ij}{}^f H_i s_j$. Along any direction in an isotropic crystal or along the optic axis in anisotropic crystals $v_1 = v_2$ and the two ellipses, degenerate into right and left circularly polarised waves travelling with refractive indices n_r and n_l respectively. The rotatory power is given by

$$\rho = \frac{\pi}{\lambda}(n_r - n_l)$$

$$= \frac{\pi}{\lambda} n^3{}_m \gamma \tag{7}$$

where

$$n_m = \frac{n_r + n_l}{2}$$

we can write (7) in the extended form as

$$\rho = \frac{\pi}{\lambda} n^3{}_m \{\Gamma_{11}{}^f H_1 s_1 + \Gamma_{22}{}^f H_2 s_2 + \Gamma_{33}{}^f H_3 s_3 + \Gamma_{12}{}^f H_1 s_2 + \Gamma_{21}{}^f H_2 s_1 + \Gamma_{23}{}^f H_2 s_3 + \Gamma_{32}{}^f H_3 s_2 + \Gamma_{31}{}^f H_3 s_1 + \Gamma_{13}{}^f H_1 s_3\}. \tag{8}$$

Equation (8) can be written in the following form also.

$$\rho = f_{ij} H_i s_j \tag{9}$$

where

$$(f_{ij}) = \frac{\pi}{\lambda} n^3{}_m (\Gamma_{ij}{}^f). \tag{10}$$

The tensor (f) represents the Faraday rotation in the crystal. As ρ is an axial scalar, H_i an axial vector and s_i a polar vector, (f) is a second rank polar tensor, which however is not symmetric as (Γ) is itself not symmetric. The forms of the matrix (f) in the various point groups are given in Table I, together with the number of independent components.

3. Transverse Faraday Rotation

From the forms of the Faraday rotation matrices (f) we see clearly that in uniaxial crystals belong to the Group A (Table. I) and lower symmetries, the Faraday rotation is observable not only along the field direction but also in a direction transverse to it. The second effect which may be called the transverse Faraday effect may be effectively used along with the familiar longitudinal Faraday effect to extract the components of the matrix (f).

TABLE I

Forms of (f) *matrices in different point groups*

Triclinic 1, $\bar{1}$	$\begin{bmatrix} f_{11} & f_{12} & f_{13} \\ f_{21} & f_{22} & f_{23} \\ f_{31} & f_{32} & f_{33} \end{bmatrix}$	(9)
Monoclinic 2, *m*, 2/*m*	$\begin{bmatrix} f_{11} & 0 & f_{13} \\ 0 & f_{22} & 0 \\ f_{31} & 0 & f_{33} \end{bmatrix}$	(5)
Orthorhombic 222, *mm*2, *mmm*,	$\begin{bmatrix} f_{11} & 0 & 0 \\ 0 & f_{22} & 0 \\ 0 & 0 & f_{33} \end{bmatrix}$	(3)
Uniaxial Group A—3, $\bar{3}$, 4, $\bar{4}$, 4/*m*, 6, $\bar{6}$, 6/*m*	$\begin{bmatrix} f_{11} & f_{12} & 0 \\ -f_{12} & f_{11} & 0 \\ 0 & 0 & f_{33} \end{bmatrix}$	(3)
Group B—32, 3*m*, $\bar{3}m$, 422, 4*mm*, $\bar{4}2m$, 4/*mmm*, 622, 6*mm*, $\bar{6}m2$, 6/*mmm*	$\begin{bmatrix} f_{11} & 0 & 0 \\ 0 & f_{11} & 0 \\ 0 & 0 & f_{33} \end{bmatrix}$	(2)
Cubic 23, *m*3, 432, $\bar{4}3m$, *m*3*m*	$\begin{bmatrix} f_{11} & 0 & 0 \\ 0 & f_{11} & 0 \\ 0 & 0 & f_{11} \end{bmatrix}$	(1)

To observe this interesting effect one should measure rotation in a direction normal to **H**. Transverse rotation cannot be observed in uniaxial crystals belonging to Group A along the optic axis. Method of observing this in lower symmetry classes like one of orthorhombic symmetry is given below.

There are three independent constants f_{11}, f_{22} and f_{33}. Rotation per unit length ρ in any general direction s is given by

$$\rho = f_{11}H_1s_1 + f_{22}H_2s_2 + f_{33}H_3s_3$$

If s_1', s_2', and s_3' are the direction cosines of the field **H**, the above equation becomes

$$\rho = (f_{11}s_1' s_1 + f_{22}s_2' s_2 + f_{33}s_3' s_3) \mathrm{H}. \tag{11}$$

As observations are to be made along the optic axis, whose positions are fixed with respect to the index ellipsoid, the components of *s* and *s'* will be taken with respect to the principal axes X_1, X_2 and X_3 of the index ellipsoid.

In orthorhombic crystals the principal axes of the index ellipsoid coincide with the three crystallographic directions ***a***, ***b*** and ***c***. If $n_1 > n_2 > n_3$, the two optic axes will lie on the (X_1, X_3) plane. Let OP_1 and OP_2 be the two optic axes each making an angle V with the X_3 axis with 2V as the optic axial angle. Hence the rotations ρ_1 and ρ_2 along OP_1 and OP_2 for the same **H** are given by

$$\left.\begin{aligned} \rho_1 &= (f_{11}s_1' \sin \mathrm{V} + f_{33}s_3' \cos \mathrm{V}) \mathrm{H} \\ \rho_2 &= (-f_{11}s_1' \sin \mathrm{V} + f_{33}s_3' \cos \mathrm{V}) \mathrm{H} \end{aligned}\right\}. \tag{12}$$

If observations are made along one of the axes, say OP_1, and the direction of the field is perpendicular to OP_1 and at the same time it lies on the axial plane $(X_1 X_3)$ we get the rotation

$$\rho_1^{\mathrm{T}} = (f_{11} - f_{33}) \mathrm{H} \sin \mathrm{V} \cos \mathrm{V} \tag{13}$$

which does not vanish, showing that the transverse Faraday rotation ρ_1^{T} can be observed. When the field **H** is reversed the sign of ρ_1^{T} also changes.

Again when the field **H** [which is normal to OP_1] is normal to the axial plane, [*i.e.*, $s_1' = {}'_3s = 0$ and $s_2' = 1$] we find $\rho_1^{\mathrm{T}} = 0$. Thus as the crystal is rotated about the optic axis with **H** remaining normal to it, rotation changes from a maximum value of $+\rho_1^{\mathrm{T}}$ to a minimum value of $-\rho_1^{\mathrm{T}}$ and then back to $+\rho_1^{\mathrm{T}}$.

4. EXTRACTION OF COMPONENTS

By measuring longitudinal and transverse Faraday rotations along the optic axis one can get a number of tensor components. Methods of obtaining the various tensor components in different classes are given below.

(i) *Cubic crystals*.—Faraday rotation in any direction *s* for a field **H** is given by

$$\rho = f_{11} (s_1' s_1 + s_2' s_2 + s_3' s_3) \mathbf{H} \tag{14}$$

where s' is the direction of the magnetic field. We can write (14) as

$$\rho = f_{11}\, H \cos\phi$$

where ϕ is the angle between **H** and s. The rotation ρ is a maximum when $\phi = 0$, which is the longitudinal effect. The rotation ρ^L is given by

$$\rho^L = f_{11}\, H. \tag{15}$$

From this relation we get f_{11}.

(ii) *Uniaxial crystals*:

(α) *Crystals belonging to Group A.*—Faraday rotation in any general direction is given by

$$\rho = [f_{11}(s_1' s_1 + s_2' s_2) + f_{33} s_3' s_3 + f_{12}(s_1' s_2 - s_2' s_1)]\, H. \tag{16}$$

As said earlier transverse rotation is absent along the optic axis. The longitudinal rotation along the optic axis is given by

$$\rho_1^L = f_{33}\, H.$$

Hence only the constant f_{33} can be obtained.

(β) *Crystals belonging to Group* (B)—Faraday rotation in any genera direction s is given by

$$\rho = [f_{11}(s_1' s_1 + s_2' s_2) + f_{33} s_3' s_3]\, H. \tag{17}$$

The longitudinal rotation along the optic axis is

$$\rho^L = f_{33}\, H \tag{18}$$

from which f_{33} can be obtained.

(iii) *Orthorohombic crystals.*—Equation (12) gives Faraday rotation along the two optic axes for the same field **H**. Longitudinal rotation along the two axes will be identical given by

$$\rho_1^L = \rho_2^L = (f_{11} \sin^2 V + f_{33} \cos^2 V)\, H. \tag{19}$$

Equation (13) which describes transverse rotation gives one more equation in f_{11} and f_{33}. These two equations can be solved to get f_{11} and f_{33}. Thus 2 out of 3 unknowns can be obtained easily.

(iv) *Monoclinic crystals.*—There are 5 unknowns to determine and Faraday rotation in direction s is given by

$$\rho = (f_{11}s_1' s_1 + f_{22}s_2' s_2 + f_{33}s_3' s_3 + f_{13}s_1' s_3 + f_{31}s_3' s_1) \mathrm{H}.$$

Two cases must be considered.

(α) *The case when symmetry element relates the two optic axes.*—If the two optic axes OP_1 and OP_2 (with 2V as the angle between them) lie on the (X_2 X_3) plane (we are referring to the same orthogonal system that was considered in Section 3), then the crystallographic diad which is along X_2 will relate the two optic axes. Rotations ρ_1 and ρ_2 along the two optic axes are given by

$$\rho_1 = (f_{22}s_2' \sin \mathrm{V} + f_{33}s_3' \cos \mathrm{V} + f_{13}s_1' \cos \mathrm{V}) \mathrm{H} \tag{20}$$

and

$$\rho_2 = (-f_{22} s_2' \sin \mathrm{V} + f_{33} s_3' \cos \mathrm{V} + f_{13} s_1' \cos \mathrm{V}) \mathrm{H}. \tag{21}$$

Longitudinal rotations along the two axes are the same given by

$$\rho_1{}^{\mathrm{L}} = \rho_2{}^{\mathrm{L}} = (f_{22} \sin^2 \mathrm{V} + f_{33} \cos^2 \mathrm{V}) \mathrm{H}. \tag{22}$$

For the transverse rotation we can have two possibilities. If observations are made along OP_1 and the transverse field **H** lies on the axial plane we get

$$\rho_1{}^{\mathrm{T}} = (f_{22} - f_{33}) \mathrm{H} \sin \mathrm{V} \cos \mathrm{V}. \tag{23}$$

However if the transverse field is normal to the axial plane

$$\rho_1{}^{\mathrm{T}} = f_{13} \mathrm{H}. \tag{24}$$

Hence using (22), (23) and (24) we get three constants f_{22}, f_{33} and f_{13}.

(β) *The case when the symmetry element does not relate the two optic axes.*—If OP_1 and OP_2 lie on the (X_1X_3) plane the diad which is along X_2 does not relate the two optic axes. Rotations ρ_1 and ρ_2 along the two optic axes are given by

$$\rho_1 = (f_{11}s_1' \sin \mathrm{V} + f_{33}s_3' \cos \mathrm{V} + f_{13}s_1' \cos \mathrm{V} + f_{31}s_3' \sin \mathrm{V}) \mathrm{H}$$

and

$$\rho_2 = (-f_{11}s_1' \sin \mathrm{V} + f_{33}s_3' \cos \mathrm{V} + f_{13}s_1' \cos \mathrm{V} - f_{31}s_3' \sin \mathrm{V}) \mathrm{H}. \tag{25}$$

If ρ_1^L and ρ_2^L are the longitudinal rotations along OP_1 and OP_2, then we get

$$\rho_1^L = [f_{11} \sin^2 V + f_{33} \cos^2 V + (f_{13} + f_{31}) \sin V \cos V] H \tag{26}$$

and

$$\rho_2^L = [f_{11} \sin^2 V + f_{33} \cos^2 V - (f_{13} + f_{31}) \sin V \cos V] H. \tag{27}$$

If the crystal is placed such that **H** is normal to OP_1 and at the same time lies also on the axial plane, then the transverse rotation along OP_1 is given by

$$\rho_1^T = [(f_{33} - f_{11}) \sin V \cos V - f_{13} \cos^2 V + f_{31} \sin^2 V] H. \tag{28}$$

A similar experiment along OP_2 gives

$$\rho_2^T = [(f_{33} - f_{11}) \sin V \cos V + f_{13} \cos^2 V + f_{31} \sin^2 V] H. \tag{29}$$

Hence using (26), (27), (28) and (29) we get 4 out of 5 constants. We have considered only the symmetry element of the diad. The case of mirror as a symmetry element can be easily worked out. In the point group $2/m$ the mirror is perpendicular to the diad. In this case the optic axes will be symmetrically related and equations from (20) to (24) can be used. For crystals belonging to point group 'm' we have only mirror symmetry. If 'm' coincides with the $(X_1\ X_2)$ plane and the two optic axes are in the $(X_2\ X_3)$ plane then they will be symmetrically related and equations from (20) to (24) can be used. On the other hand if the two axes are on the $(X_1\ X_2)$ plane itself then they will not be related through the symmetry element.

(v) *Triclinic crystals.*—There are 9 unknowns in this case and the index ellipsoid is disposed with its principal axes at a general orientation with respect to the crystallographic directions. As in the previous cases here also we refer components of s and **H** with respect to the principal axes of the index ellipsoid. If the two optic axes lie on the $(X_1\ X_2)$ plane each at an angle V from the X_3 axis, then the magneto-optic rotations along the two optic axes will be

$$\rho_1 = (f_{11}s_1' \sin V + f_{33}s_3' \cos V + f_{13}s_1' \cos V + f_{31}s_3' \sin V$$
$$+ f_{23}s_2' \cos V + f_{21}s_2' \sin V) H \tag{30}$$

and

$$\rho_2 = (-f_{11}s_1' \sin V + f_{33}s_3' \cos V + f_{13}s_1' \cos V - f_{31}s_3' \sin V + f_{23}s_2' \cos V - f_{21}s_2' \sin V)\, H.$$

Longitudinal rotations $\rho_1{}^L$ and $\rho_2{}^L$ along the two optic axes will be not equal. They are given by

$$\rho_1{}^L = [f_{11} \sin^2 V + f_{33} \cos^2 V + (f_{13} + f_{31}) \sin V \cos V]\, H \tag{31}$$

and

$$\rho_1{}^L = [f_{11} \sin^2 V + f_{33} \cos^2 V - (f_{13} + f_{31}) \sin V \cos V]\, H. \tag{32}$$

Transverse rotations along the two optic for **H** normal to the axial plane are given by

$$\rho_1{}^T = (f_{22} \cos V + f_{21} \sin V)\, H \tag{33}$$

and

$$\rho_2{}^T = (f_{22} \cos V - f_{21} \sin V)\, H. \tag{34}$$

Transverse rotations $\rho_1{}^{T'}$ and $\rho_2{}^{T'}$ when **H** lies on the axial plane are given by

$$\rho_1{}^{T'} = [(f_{33} - f_{11}) \sin V \cos V - f_{13} \cos^2 V + f_{31} \sin^2 V]\, H \tag{35}$$

and

$$\rho_2{}^{T'} = [(f_{33} - f_{11}) \sin V \cos V + f_{13} \cos^2 V - f_{31} \sin^2 V]\, H. \tag{36}$$

Thus we have 6 independent equations from which we can get f_{11}, f_{33}, f_{13}, f_{31}, f_{22} and f_{21}. Hence 6 out of 9 unknowns can be obtained.

5. Faraday Rotation in Polycrystals

It is possible to make a randomly oriented crystalline aggregate of anisotropic crystals. The medium as a whole will be translucent if optically anisotrophy of the crystallites are large. On the other hand one can get a good amount of transmission for weakly birefringent crystals. If the crystallites are optically inactive the medium will behave as an optically isotropic media. A plane polarized light emerges out of the medium as partially polarized light. The completely polarized part of the emergent beam will be in the same state of polarization as the incident (Ramaseshan, (1972).

In the presence of a magnetic field each crystallite will become magnetically active resulting in Faraday rotation. Under these conditions the medium will still be optically isotropic exhibiting a Faraday rotation whose magnitude is

$$\bar{\rho} = \bar{f}\mathrm{H} \tag{37}$$

where

$$\bar{f} = \tfrac{1}{3}(f_{11} + f_{22} + f_{33}) \tag{38}$$

with f_{11}. f_{22}, f_{33} as the principal components of (f). Also there will be a slight increase in depolarization. In all the crystal classes considered so far we could get only two out of three principal components. It follows from (27) and (28) that by measuring Faraday rotation in the polycrystal we can get $\bar{f}$ from which the remaining principal component can also be obtained. Hence one more constant can be obtained in all the non-cubic classes. Table II gives the number of components of (f) matrix that can be obtained from single and polycrystal data.

TABLE II

Crystal		No. of components obtainable		
	No. of unknowns	Single crystal	Poly-crystal	Total
Triclinic ..	9	6	1	7
Monoclinic ..	5	4 3	1 1	5 4
Orthorhombic ..	3	2	1	3
Uniaxial				
Group A ...	3	1	1	2
Group B ..	2	1	1	2
Cubic ..	1	1	...	1

6. PIEZO-FARADAY EFFECT

In recent times stress has been found to alter Faraday rotation in crsytals [Skaggs and Broersma (1964)]. This problem has been theoretically worked out by Bhagavantam (1971) who has studied the effect of stress on the optical properties of magnetic crystals. Magnetic crystals in general show Faraday type of rotation. However in certain magnetic symmetry point groups this phenomenon will be absent. Bhagavantam has found an interesting result that even these magnetic point groups, which normally forbid Faraday type rotation, show up magnetic rotation under the influence of stress. In cubic crystals belonging to $m3$ and $m3\underline{m}$ this effect has been predicted. It would be interesting to workout methods of observing these effects which will not be straightforward due to the unavoidable photoelastic effects, that accompany stresses.

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