# ON THE PHENOMENOLOGICAL THEORY OF LIGHT PROPAGATION IN OPTICALLY ACTIVE CRYSTALS

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## 1. INTRODUCTION

THE treatment customarily adopted[1] for analysing the propagation of light in optically active crystals is somewhat lengthy and cumbersome, and is even so not free from approximation: its mathematical complexity being perhaps partly the reason why an alternative quasi-theoretical analysis not involving the electromagnetic theory is sometimes used.[2,3] A concise yet logical treatment of the problem is, however, easily possible without the introduction of any fresh physical hypothesis—as will be shown in this paper.

We have merely to remember that for an electromagnetic wave inside a crystal, it is the dielectric displacement vector **D** (the 'vibration'), and not the electric intensity **E** which necessarily lies on the wave-front. Hence we seek to express **E** in terms of **D**, rather than *vice-versa*. Even for the case of optically inactive crystals, such a procedure—though not commonly adopted—is known to simplify the theoretical treatment, as may be seen especially in the case of absorbing crystals[4]; and the geometric representation of the optical properties of a crystal by an *index ellipsoid* follows directly in the structure of such a presentation instead of having to be proved by indirect means later—as is usually done.[5]

## 2. FORMULATION OF THE PROBLEM

Any wave travelling in the crystal along an arbitrary direction '$z$' (conveniently taken as being normal to the plane of the paper) may be specified by giving the $x$ and $y$ components of the D-vibration: $D_x \exp i\omega (t - z/v)$ and $D_y \exp i\omega (t - z/v)$. Here $v$ is the velocity of the wave and the ratio $(D_y/D_x)$ completely defines the polarisation of the wave. Owing to the requirement that the vectors **D** and **E** must satisfy both Maxwell's relations as well as the characteristic polarisable property of the medium, only two specific waves can be propagated in the $z$-direction—the problem being to determine their velocities, $v_1$ and $v_2$, and their states of polarisation $(D_y/D_x)_1$ and $(D_y/D_x)_2$.

Maxwell's equations for a plane-wave field reduce to the following single equation when **H** is eliminated*—as is shown in standard texts[7]

$$v^2\mathbf{D} = c^2 [\mathbf{E} - \mathbf{s} (\mathbf{E}.\mathbf{s})] \tag{1}$$

where **s** is a unit-vector along the wave-normal. For inactive crystals the polarisable characteristics of the medium lead to the usual relation $\mathbf{D} = (\epsilon)\,\mathbf{E}$, where $(\epsilon)$ represents the symmetric dielectric tensor—which operates on **E** to give **D**. But for optically active media it is assumed that

$$\mathbf{D} = (\epsilon)\,\mathbf{E} + i\,\mathbf{G}\times\mathbf{E} \tag{2}$$

where the additional antisymmetric term represents a feeble additional polarisation which oscillates out of phase with the electric intensity (as indicated by the factor $i$), and yet does not introduce any absorption because it is also orthogonal to the electric intensity (as is indicated by the occurrence of the vector product). The *gyration-vector* **G** corresponding to the direction of propagation **s** determines the optical rotatory power in that direction.

## 3. The Inverse Gyration-Vector

It is known that by taking the $z$-axis along the wave-normal (instead of choosing co-ordinate axes along the electrical axes, as in usual treatments), we not only confine ourselves to two components of **D** instead of three; in addition, the components of the vector equation (1) take the following intelligible form:

$$v^2\mathrm{D}_x = c^2\mathrm{E}_x;\;\; v^2\mathrm{D}_y = c^2\mathrm{E}_y;\;\; D_z = 0 \tag{1 a}$$

where, it may be noted, separate and explicit expressions are obtained for $\mathrm{E}_x$ and $\mathrm{E}_y$.

Equation (2) which expresses **D** as a vector function of **E**, is now transformed by expressing **E** in terms of **D** using the inverse vector function. Since this inverse function will obviously contain not only a real symmetric part but also an imaginary antisymmetric part (giving that part of the electric intensity which oscillates out of phase with **D**, and is orthogonal to it), we may write

$$c^2\mathbf{E} = (a)\,\mathbf{D} - i\,\Gamma\times\mathbf{D} \tag{2 a}$$

The first term alone would be present for an optically inactive crystal, the symmetric tensor $(a)$ then defining the *index ellipsoid*†—being proportional to the inverse of the dielectric tensor (see Appendix). The comparatively

* We are not here considering other theories of optical activity[2,6] in which $\mathrm{B} \neq \mathrm{H}$.

† If $a_{11}x^2 + a_{22}y^2 + \ldots .\, 2a_{31}\,zx = 1$, be the equation to the index ellipsoid, then the $a_{ij}$ are also the components of the tensor $(a)$ with respect to the co-ordinate axes chosen.

small second term (which introduces the optical activity) is determined by the new vector $\Gamma$—which we shall refer to as the inverse gyration-vector corresponding to the direction of propagation **s**.

## 4. Derivation of the Equations of Wave-Propagation

Writing down the $x$ and $y$ components of equation (2 $a$) and omitting the terms containing the factor $D_z$ (since $D_z = 0$):

$$\left.\begin{aligned} c^2E_x &= a_{11}D_x + a_{12}D_y + i\Gamma_z D_y \\ c^2E_y &= a_{22}D_y + a_{12}D_x - i\Gamma_z D_x \end{aligned}\right\} \tag{3}$$

Here $a_{11}x^2 + a_{22}y^2 + 2a_{12}xy = 1$, is the equation to the elliptical section of the index ellipsoid taken perpendicular to the wave-normal. We now choose the $x$ and $y$ axes parallel to the principal radii of this elliptical section, which represent the vibration-directions in the absence of optical activity. *With this understanding* we may, in Equation (3), set $a_{12} = 0$, $a_{11} = v'^2$ and $a_{22} = v''^2$; here $v'$ and $v''$ represent the velocities of the waves in the absence of optical activity, because of a well-known property of the index ellipsoid which may also be obtained by setting $\Gamma_z = 0$ at the end of the present discussion. Substituting for $c^2E_x$ and $c^2E_y$ from equation (1 $a$) we thus obtain as our fundamental equations:

$$\left.\begin{aligned} v^2 - v'^2 &= i\Gamma_z\left(\frac{D_y}{D_x}\right) \\ v^2 - v''^2 &= -\, i\Gamma_z\left(\frac{D_x}{D_y}\right) \end{aligned}\right\} \tag{4}$$

These fundamental equations are essentially the same as those obtained in Pockel's *Lehrbuch*[2] starting from an entirely different theory of optical activity. There are two pairs of roots $v_1$, $(D_y/D_x)_1$ and $v_2$, $(D_y/D_x)_2$, which simultaneously satisfy these equations—giving thereby the required velocities and states of polarisation.

## 5. The Velocities and States of Polarisation of the Waves

Multiplying the two equations of (4) to eliminate $(D_y/D_x)$ we obtain the following quadratic in $v^2$, whose roots $v_1^2$ and $v_2^2$ give the velocities of the waves:

$$(v^2 - v'^2)(v^2 - v''^2) = \Gamma_z^2 \tag{5}$$

Subtracting the second equation of (4) from the first to eliminate $v^2$, we get:

$$\frac{D_y}{D_x} + \frac{D_x}{D_y} = i\,\frac{v'^2 - v''^2}{\Gamma_z} \tag{6}$$

This is a quadratic in $(D_y/D_x)$, and its roots uniquely determine the states of polarisation because of the relation:

$$\frac{D_y}{D_x} = \frac{|D_y|}{|D_x|} e^{iR}$$

where the ratio $|D_y| : |D_x|$ is obviously the ratio $\tan\theta$ of the (real) amplitudes of the $y$ and $x$ components of the D-vibration; and R is the relative phase difference between these components. Since the right-hand side of (6) is entirely imaginary it follows that both roots of the equation are imaginary ($R = \pm \pi/2$). Further since the roots of (6) are obviously reciprocals of one another, it follows that if one root is of the form $(B_1/A_1)\, e^{i\pi/2}$, the other will be $(A_1/B_1)\, e^{-i\pi/2}$. This signifies that both vibrations are elliptically polarised with their respective major axes along the two perpendicular principal planes, the vibrations being of the same ellipticity but traced in opposite senses.

The value of $\tan 2\theta$, which determines the common ellipticity of the two vibrations is obtained directly from equation (6) by substituting in it the value of $(D_y/D_x)$ which is equal to $(+\, i \tan\theta_1)$ for the right elliptic vibration and $(-\, i \tan\theta_2)$ for the left elliptic vibration:

$$|\tan 2\theta| = |2\Gamma_z / (v'^2 - v''^2)| \tag{7}$$

Similarly the difference in the squares of the velocities of the two waves may be obtained from (5)—since it is the square-root of the discriminant:

$$(v_1^2 - v_2^2) = (v'^2 - v''^2) + (2\Gamma_z)^2 \tag{8}$$

In the absence of linear birefringence (*i.e.*, if we imagine $v' = v''$), the waves will be circularly polarised according to (7), the rotatory power $\rho$ being obtained from (8) as:

$$\rho = (\omega / v_m^3) \cdot \Gamma_z \tag{9}$$

where $v_m$ is a mean velocity (see Pockels[2]).

We may mention that because of an approximation made in the usual treatment, its resultant equations differ slightly from (5) and (6); also, equations (7) and (8) are the accurate forms of the approximate relations obtained by the method of superposition.[2,3]

## 6. The Variation of Rotatory Power with Direction

The gyration vector **G**, and hence in the present treatment the inverse gyration-vector $\Gamma$, are both vectors not having their axes fixed with respect to the crystallographic axes,‡ but are linear vector functions of the direction of propagation. Thus $\mathbf{G} = (g)\,\mathbf{s}$ and $\Gamma = (\gamma)\,\mathbf{s}$, where the gyration tensor $(g)$ and hence the modified gyration tensor $(\gamma)$ are both nine-component tensors. The rotatory power is however determined, according to (9), only by the resolved component $\Gamma_S$ of the inverse gyration-vector along the direction of propagation. (We are here denoting this by $\Gamma_S$ instead of $\Gamma_Z$ since we are choosing an arbitrary co-ordinate system $x'y'z'$.) Since an analogous result holds in the usual treatment it follows as in that case[6] that this scalar parameter of rotation $\Gamma_S$ is a quadratic function of the direction cosines $l'm'n'$ of propagation:

$$\Gamma_S = \gamma_{11}l'^2 + \gamma_{22}m'^2 + \gamma_{33}n'^2 + 2\bar{\gamma}_{12}l'm' + 2\bar{\gamma}_{23}m'n' + 2\bar{\gamma}_{31}n'l'$$

where

$$\bar{\gamma}_{ij} = \tfrac{1}{2}(\gamma_{ij} + \gamma_{ji}), \qquad (i, j, = 1, 2, 3).$$

The discussion of optical activity in relation to crystal symmetry follows as in the usual treatment.

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## 7. Summary

In the usual treatment of the optical activity of crystals, the displacement vector of the light wave is expressed as a function of the electric vector—this being done with the aid of the dielectric tensor and the gyration-vector. The treatment is made more concise and tractable by using the inverse vector function and expressing the electric vector in terms of the displacement vector, since it is the latter which necessarily lies on the wave-front; this can be done by using the inverse of the dielectric tensor (which defines the index ellipsoid), and an inverse gyration-vector (which determines the rotatory power for the particular direction of propagation considered). The inverse gyration-vector (like the gyration-vector) is itself a function of the direction of propagation, being related to it by a modified gyration tensor—which consequently determines the rotatory power for all directions.

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‡ The new treatment given in Sommerfeld's *Optics*[5] is deficient in this respect.

## 8. REFERENCES


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## APPENDIX

Since the index ellipsoid defined by the tensor $(a)$, and the inverse gyration-vector $\Gamma$ may be directly taken as the phenomenological quantities required to describe the propagation along any particular direction **s**, the relation between $\Gamma$ and the gyration-vector **G** would not usually be required. The relation may however be obtained by writing (2) as $\mathbf{D}=\mathbf{D}' + i\mathbf{P}''$, where

$$\mathbf{D}' = (\epsilon)\,\mathbf{E};\;\; \mathbf{P}'' = \mathbf{G}\times\mathbf{E} \qquad (11)$$

Substituting for **D** in (2 *a*)

$$c^2\mathbf{E} = (a)\,\mathbf{D}' + \Gamma\times\mathbf{P}'' + i\,(a)\mathbf{P}'' - i\Gamma\times\mathbf{D}'$$

Equating real parts and neglecting $\Gamma\times\mathbf{P}''$ in comparison with $c^2\mathbf{E}$:

$$(a) = c^2\,(\epsilon)^{-1} \qquad (12)$$

Equating imaginary parts: $(a)\,\mathbf{P}'' = \Gamma\times\mathbf{D}'$. Choosing axes of co-ordinates X, Y, Z, along the principal electrical axes of the crystal, the X-component of this relation is

$$a_X P_X'' = \Gamma_Y D_Z' - \Gamma_Z D_Y'$$

or, by virtue of (11), if $\epsilon_X$, $\epsilon_Y$, $\epsilon_Z$, be the principal dielectric constants,

$$a_X\,(G_Y E_Z - G_Z E_Y) = \Gamma_Y\,(\epsilon_Z E_Z) - \Gamma_Z\,(\epsilon_Y E_Y)$$

We may separately equate the coefficients of $E_Y$ and $E_Z$ occurring on both sides since this relation has to hold not just for *one* value of the ratio $E_Y/E_Z$, there being *two* polarised waves that can be propagated along the same direction. Hence, using (12),

$$\Gamma_Y = \frac{c^2}{\epsilon_Z\epsilon_X} G_Y;\;\; \Gamma_Z = \frac{c^2}{\epsilon_X\epsilon_Y}\cdot G_Z;\;\; \Gamma_X = \frac{c^2}{\epsilon_Y\epsilon_Z}\cdot G_X \qquad (13)$$

The relation between the components of the modified gyration tensor and those of the gyration tensor may now be easily written down:

$$\gamma_{1i} = \frac{c^2}{\epsilon_Y\epsilon_Z} g_{1i};\;\; \gamma_{2i} = \frac{c^2}{\epsilon_Z\epsilon_X} g_{2i};\;\; \gamma_{3i} = \frac{c^2}{\epsilon_X\epsilon_Y} g_{3i} \qquad (i = 1, 2, 3).$$