LIGHT PROPAGATION IN ABSORBING CRYSTALS POSSESSING OPTICAL ACTIVITY—ELECTROMAGNETIC THEORY

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§ 1. Introduction

In the present paper we shall give the electromagnetic theory of light propagation in absorbing crystals possessing optical activity: the theoretical presentation is a straightforward extension of that previously adopted for transparent active crystals.[1] We shall find that the results of the electromagnetic theory are, for most practical purposes, the same as obtained previously[2] on the basis of a simpler physical idea, *viz.*, by a 'method of superposition'—the results of which have been confirmed by detailed observations on the interesting phenomena displayed by amethyst.[3]

A complete solution of the electromagnetic theory of light propagation in absorbing active crystals has thus far been presented only for the case of uniaxial media.[4] Recently an entirely new and general method of formally solving the electromagnetic equations has been introduced by Jones[5] in a paper dealing with light propagation in anisotropic media (*see*, however, Appendix). We shall find in § 11, that it becomes essential to adopt this new method for solving the propagation of light along certain remarkable directions, *viz.*, the so-called *singular axes* which can exist both in active[3] as well as inactive[6,7] crystals.

§ 2. Formulation of the Problem

Consider an arbitrary direction of propagation Oz in the crystal—which direction we may conveniently take as being normal to the plane of the paper. Our problem is merely to determine the characteristic states of polarisation of the plane waves that can be propagated along this direction, as well as their velocities and absorption coefficients. The reason why only specific types of waves can be propagated along the z-direction is that the field vectors (in particular, the vectors **D** and **E**) of the electromagnetic wave are constrained to satisfy certain relations amongst themselves. Firstly they must satisfy Maxwell's equations; and secondly they must obey certain constitutive relations imposed by the properties of the medium, and which determine the optical characteristics of the medium. These latter relations will be determined by the polarisable characteristics of the medium, *i.e.*, the relation which the induced polarisation bears to the electric field of the light wave. (As is customary even in the case of transparent active crystals we shall assume for simplicity that there is no induced magnetization, *i.e.*, that $\mathbf{B} = \mathbf{H}$, though—as in that case—this assumption appears to violate energy considerations.)

§ 3. Maxwell's Relations for a Plane Wave Field

Since we are interested in plane waves propagating in the z-direction, the field vectors do not vary over planes normal to the z-direction. If $\mathbf{k}$ denote a unit vector along the z-axis, we may use the operator $\mathbf{k}(\partial/\partial z)$ in place of the gradient operator ∇ in the Maxwell's equations—or rather in the standard relation obtained by eliminating $\mathbf{H}$ between the Maxwell's equations. The latter relations (*see*, *e.g.*, Page and Adams,[8] eq. 82–9) then assume the form

$$\ddot{\mathbf{D}} = c^2 \frac{\partial^2}{\partial z^2} [\mathbf{E} - \mathbf{k}(\mathbf{k} \cdot \mathbf{E})]. \tag{1}$$

The components of this equation take the simpler form

$$\ddot{D}_x = c^2 \frac{\partial^2 E_x}{\partial z^2}; \qquad \ddot{D}_y = c^2 \frac{\partial^2 E_y}{\partial z^2} \tag{2}$$

$$\ddot{D}_z = 0. \tag{3}$$

The last condition implies the transversality of the displacement vector—since we shall be concerned only with fields varying harmonically in time. Also we shall first restrict our consideration to homogeneously polarised plane waves, so that

$$\mathbf{D}, \mathbf{E} \sim \exp.\ i\omega\left(t - \frac{z}{c} \cdot \bar{n}\right) \times \mathbf{D}_0, \mathbf{E}_0 \tag{4}$$

where n is the complex refractive index. Under these conditions the operation $\partial/\partial z$ becomes identical with multiplication by $-i\omega\bar{n}/c$ and the operation $\partial/\partial t$ with multiplication by $i\omega$. Thus for a plane wave field of type (4) Maxwell's equations (with $\mathbf{H}$ eliminated) finally assume the elegant form

$$\bar{v}^2 D_x = c^2 E_x; \qquad \bar{v}^2 D_y = c^2 E_y \tag{5}$$

where $\bar{v}$ represents the complex velocity $c/\bar{n}$ of the damped wave.

§ 4. PROPERTIES OF THE MEDIUM

The dielectric displacement in the medium depends on the electric vector, and, for a transparent optically active crystal,[9] **D** may be expressed as an explicit vector function of **E** by the use of the dielectric tensor (ϵ) and gyration tensor (g); it was shown in a previous paper[1] that it was more convenient to reverse the procedure and express **E** as an explicit function of **D** by the use of the index-tensor (a), and a modified gyration tensor (γ). When we turn to media possessing absorption, the components of the dielectric tensor and the gyration tensor will become complex quantities: in terms of our modified presentation it may be shown that the consequence of this is that the constants of the index tensor and the modified gyration tensor have to be replaced by complex quantities (*see* Appendix). Thus the relation between **E** and **D** may be expressed in the same form as previously given for transparent active crystals (reference 1, eq. 2 *a*).

$$c^2\mathbf{E} = (\bar{a})\,\mathbf{D} - i\bar{\Gamma} \times \mathbf{D}. \tag{6}$$

Here ($\bar{a}$) is a symmetric tensor—the complex index-tensor; and $\bar{\Gamma}$ is an auxiliary vector which depends on the direction of the wave-normal **s**, being given by

$$\bar{\Gamma} = (\bar{\gamma})\,\mathbf{s} \tag{7}$$

where ($\bar{\gamma}$) represents the 'tensor of optical activity'—a general tensor with complex components. It is convenient to express the above relations in terms of tensors having real components; *these tensors will in turn separately determine the various optical characteristics of the medium.*

Thus in (6) and (7) we may substitute

$$(\bar{a}) = (a) + i\,(b) \tag{8}$$

$$(\gamma) = (\gamma) + i\,(\gamma'). \tag{9}$$

Here (a) and (b) are the usual index- and absorption-tensors which occur for example in optically inactive absorbing crystals, and which define the *index-* and *absorption-ellipsoids*; (γ) is the tensor of optical rotation which was referred to in our paper on transparent active crystals[1] as the modified gyration tensor. The new tensor (γ') may be called the tensor of circular dichroism for reasons which will become apparent as we proceed. In eq. (6) it is clear that we may further substitute

$$\bar{\Gamma} = \Gamma + i\Gamma' \tag{10}$$

where

$$\Gamma = (\gamma)\,\mathrm{s} \tag{11}$$

$$\Gamma' = (\gamma')\,\mathrm{s}. \tag{12}$$

§ 5. SOLUTION OF THE ELECTROMAGNETIC EQUATIONS

We now write down the x and y components of the vector equation (6) after expanding the cross-product and omitting terms in D_z (since the latter is equal to zero).

$$\left.\begin{aligned} c^2 E_x &= \bar{a}_{11} D_x + (\bar{a}_{12} + i\bar{\Gamma}_z)\, D_y \\ c^2 E_y &= (\bar{a}_{12} - i\bar{\Gamma}_z)\, D_x + \bar{a}_{22} D_y \end{aligned}\right\}. \tag{13}$$

Substituting for c^2E_x and c^2E_y from (5) we obtain as our fundamental equations:

$$\left.\begin{aligned} \bar{v}^2 - \bar{a}_{11} &= (\bar{a}_{12} + i\bar{\Gamma}_z)\left(\frac{D_y}{D_x}\right) \\ \bar{v}^2 - \bar{a}_{22} &= (\bar{a}_{12} - i\bar{\Gamma}_z)\left(\frac{D_x}{D_y}\right) \end{aligned}\right\}. \tag{14}$$

In general there will be two pairs of roots $\bar{v}_a$, $(D_y/D_x)_a$ and $\bar{v}_b$ $(D_y/D_x)_b$ which will satisfy these simultaneous equations. These will give the complex velocities and states of polarisation of the two waves that can be propagated in the z-direction. If we multiply the two equations of (14) to eliminate (D_y/D_x) we obtain the following quadratic in $\bar{v}^2$ whose roots $\bar{v}_a{}^2$ and $\bar{v}_b{}^2$ determine the complex velocities (*i.e.*, the velocities and absorption coefficients of the waves):

$$(\bar{v}^2 - \bar{a}_{11})(\bar{v}^2 - \bar{a}_{22}) = \bar{a}_{12}{}^2 + \bar{\Gamma}_z{}^2. \tag{15}$$

Subtracting the second equation of (14) from the first to eliminate $\bar{v}^2$, we get

$$(\bar{a}_{12} + i\bar{\Gamma}_z)\left(\frac{D_y}{D_x}\right) - (\bar{a}_{12} - i\bar{\Gamma}_z)\left(\frac{D_x}{D_y}\right) = -(\bar{a}_{11} - \bar{a}_{22}).$$

Or,

$$(\bar{a}_{12} + i\bar{\Gamma}_z)\left(\frac{D_y}{D_x}\right)^2 + (\bar{a}_{11} - \bar{a}_{22})\left(\frac{D_y}{D_x}\right) - (\bar{a}_{12} - i\bar{\Gamma}_z) = 0. \tag{16}$$

This is a quadratic in (D_y/D_x) and *its roots specify the states of polarisation of the waves.* Since these roots will in general be complex, the waves will be elliptically polarised.

The task of discussing in greater detail the velocities, absorption coefficients and states of polarisation of the waves is complicated by the fact that all the coefficients occurring in (15) and (16) are really complex quantities:

$$\bar{a}_{hk} = a_{hk} + ib_{hk}\ ; \quad \bar{\Gamma}_z = \Gamma_z + i\Gamma_z'. \tag{17}$$

It is worthwhile pointing out that the a_{hk} and b_{hk} (which are the components of the index and absorption tensors respectively) are also the constants occurring in the equation to the elliptic sections of the index- and absorption-ellipsoids by the xy plane; in other words the equation to these elliptic sections are respectively

$$\left.\begin{array}{l} a_{11}x^2 + a_{22}y^2 + 2a_{12}xy = 1 \\ b_{11}x^2 + b_{22}y^2 + 2b_{12}xy = 1 \end{array}\right\}. \tag{18}$$

Also Γ_z is the scalar parameter of optical rotation already met with in the case of transparent optically active crystals[1]; whilst Γ_z' may be called the scalar parameter of circular dichroism for reasons which will be discussed in the next section.

§ 6. Circular Dichroism and Its Directional Variation

The characteristic effect introduced by the presence of the parameter Γ_z' may be best revealed by supposing linear birefringence and linear dichroism to be absent, *i.e.*, by setting [in equations (15) and (16)],

$$a_{11} = a_{22} = a; \quad a_{12} = 0; \quad b_{11} = b_{22} = b; \quad b_{12} = 0$$

i.e.,

$$\bar{a}_{11} = \bar{a}_{22} = \bar{a}; \quad \bar{a}_{12} = 0.$$

Such a situation actually occurs for a direction of propagation along the uniaxial axis in a crystal of uniaxial symmetry, since the sections of the index- and absorption-ellipsoids will be circular. Equations (16) and (15) then yield

$$\frac{D_y}{D_x} = \pm i; \quad v^2 = \bar{a} \mp \bar{\Gamma}_z. \tag{19}$$

This means that the waves are right and left circularly polarised and if $\bar{v}_r$ and v_l be the complex velocities of the circularly polarised waves, then

$$\bar{v}_l^2 - \bar{v}_r^2 = 2\bar{\Gamma}_z. \tag{20}$$

The complex velocity $\bar{v}$ is related to the actual velocity v and the extinction coefficient κ by the usual relation

$$\bar{v} = \frac{c}{n - i\kappa} = v\left(1 + \frac{i\kappa v}{c}\right) \tag{21}$$

the terms containing the square of the extinction coefficient being negligible in magnitude. Introducing this in equation (20), we obtain to a high degree of approximation

$$n_r - n_l = \Gamma_z \cdot \frac{c}{v_m{}^3} = \rho_0 \tag{22}$$

$$\kappa_l - \kappa_r = \Gamma_z{}' \cdot \frac{c}{v_m{}^3} = \sigma_0 \tag{23}$$

where v_m is a mean velocity.

It will now be clear why Γ_z and $\Gamma_z{}'$ may be referred to as the parameters of optical rotation and circular dichroism corresponding to the direction of propagation z. The values of these parameters for a general direction of propagation s may be denoted by Γ_s and $\Gamma_s{}'$. The parameter of optical rotation has been shown to be a quadratic function of the direction cosines of propagation by virtue of (11) (*see* ref. 1, § 6); the same statement must therefore be true for the directional variation of the parameter of circular dichroism. If we lay off two radii vectores r_1 and r_2 parallel to the direction of propagation such that their lengths are given by

$$\frac{1}{r_1{}^2} = |\Gamma_s| = \rho_0 \cdot \frac{v_m{}^3}{c} \tag{24}$$

$$\frac{1}{r_2{}^2} = |\Gamma_s{}'| = \sigma_0 \frac{v_m{}^3}{c} \tag{25}$$

then (24) and (25) define respectively a surface of optical rotation and a surface of circular dichroism. Given these surfaces we may determine Γ_s and $\Gamma_s{}'$ for any direction of propagation, or alternatively the coefficients of circular birefringence and circular dichroism (ρ_0 and σ_0) for any direction.* (The sign to be attached may be supposed to be marked on the surface.)

§ 7. ANALYTICAL PRESENTATION OF THE METHOD OF SUPERPOSITION

The states of polarisation of the waves propagated along any direction as well as their velocities and absorption coefficients may in principle be

* We take the mean velocity v_m for the direction in question to be given by equation (14) of reference 6.

obtained from equations (15) and (16). In practice, in order to be able to study the variation of these properties with direction the results would have to be cast into a simpler parametrical form—as Voigt[10] was compelled to do even in the case of inactive absorbing crystals. Now the propagation in an absorbing active crystal has been previously analysed without using the electromagnetic theory, by a method of superposition; this method, *when developed by the use of the Poincaré sphere*, leads to results which are automatically in such a parametrical form, and we shall show in the next section that these results can be modified so as to represent accurately the results of the electromagnetic theory. Unfortunately the modifications necessary can be perceived only if the results of the method of superposition, instead of being expressed in simple parametrical form, are expressed analytically by equations formally similar to those of the electromagnetic theory. For this purpose we shall in this section develop the consequences of the superposition method not by the geometrical methods of our previous paper,[2] but by analytical methods, along the lines followed by Jones[11,12] who has adopted a matrix calculus treatment.

A detailed physical description of the method of superposition may be found in references 6 and 2. Let $\vec{D}$ be the vibration at the plane z in the crystal.† The vibration which would obtain at the plane $(z + dz)$ if the crystal were transparent and inactive may be represented by $\vec{D} + \partial_1\vec{D}$. This vibration may be obtained by multiplying the components of $\vec{D}$ along the principal planes of linear birefringence Ox' and Oy' by the factors

$$\exp.\left(-i\frac{2\pi}{\lambda_0}n_1 dz\right)$$

and

$$\exp.\left(-i\frac{2\pi}{\lambda_0}\cdot n_2 dz\right) \text{ respectively.}$$

Then

$$\left.\begin{aligned} \partial_1 D_{x'} &= -i\frac{2\pi}{\lambda_0}n_1 D_{x'} \\ \partial_1 D_{y'} &= -i\frac{2\pi}{\lambda_0}n_2 D_{y'} \end{aligned}\right\}. \qquad (26)$$

† The displacement vector is here written as a two-dimensional vector since it lies on the wavefront. Also the vibration is taken as $\vec{D}e^{i\omega t}$, *i.e.*, in the present section the time factor is not included within the symbol for the displacement vector, which is therefore only a function of z.

This may be written symbolically as

$$\partial_1 \vec{D} = - i \frac{2\pi}{\lambda_0} dz \, [n] \, \vec{D}. \tag{27}$$

where $[n]$ represents a symmetric 2×2 matrix or tensor operator whose principal directions coincide with the principal planes of linear birefringence and whose principal values are n_1 and n_2—determined from an index-ellipsoid as for a transparent inactive crsytal.

Similarly let $\partial_2 \vec{D}$ represent the partial increment to the initial vibration $\vec{D}$ when subjected to the infinitesimal operation of linear dichroism (corresponding to the thickness dz). Then we will have

$$\partial_2 \vec{D} = - \frac{2\pi}{\lambda_0} dz \, [\kappa] \, \vec{D} \tag{28}$$

where $[\kappa]$ represents a symmetric tensor whose principal directions coincide with the principal planes of linear dichroism and whose principal values are κ_1 and κ_2 as given by an absorption ellipsoid [*see* eq. (12) of ref. 6].

Under the combined effects of linear birefringence and linear dichroism the partial increment $\partial' \vec{D}$ to the initial vibration $\vec{D}$ may be obtained by adding (27) and (28). We may then write

$$\partial' \vec{D} = - i \frac{2\pi}{\lambda_0} dz \, [\bar{n}] \, \vec{D} \tag{29}$$

the components of the symmetric tensor $[\bar{n}]$ being given by

$$\bar{n}_{ij} = n_{ij} - i\kappa_{ij} \tag{30}$$

where the n_{ij} and κ_{ij} are the components of $[n]$ and $[\kappa]$ respectively.

Thus the state of vibration $\vec{D} + \partial' \vec{D}$ at the plane $z + dz$ for an *inactive* absorbing crystal is given by writing equation (29) in full:

$$\left. \begin{aligned} \partial' D_x &= - i \frac{2\pi}{\lambda_0} dz \, (\bar{n}_{11} D_x + \bar{n}_{12} D_y) \\ \partial' D_y &= - i \frac{2\pi}{\lambda_0} dz \, (\bar{n}_{12} D_x + \bar{n}_{22} D_y) \end{aligned} \right\} . \tag{31}$$

These equations correspond to eq. (29) of reference 6; the $\bar{n}_{ij}$ have the same significance as in that paper and are determined from the sections to the index- and absorption-ellipsoids.

Let $(\vec{D} + \partial_3\vec{D})$ represent the vibration obtained by subjecting the vibration $\vec{D}$ to the infinitesimal operation of rotation through an anticlockwise angle ρdz—where ρ $(\equiv \rho_0\pi/\lambda_0)$ is the optical rotatory power for the direction z as determined from a surface of optical rotation according to equation (24).

Then

$$\left.\begin{aligned} D_x + \partial_3 D_x &= D_x - \frac{\pi}{\lambda_0}\rho_0 dz . D_y \\ D_y + \partial_3 D_y &= \frac{\pi}{\lambda_0}\rho_0 dz D_x + D_y \end{aligned}\right\}. \tag{32}$$

It remains to consider the partial increment $\partial_4\vec{D}$ due to the operation of circular dichroism (corresponding to the passage dz): this operation consists in resolving the initial vibration $\vec{D}$ into its left and right circular components and multiplying the amplitudes of these components by $(1 - \pi/\lambda_0 \cdot \sigma_0 dz)$ and $(1 + \pi/\lambda_0 \cdot \sigma_0 dz)$; σ_0 is the coefficient of circular dichroism (corresponding to the difference in the extinction coefficients of the circularly polarised components) as obtained from a surface of circular dichroism according to equation (25). Clearly this operation differs from the operation of optical rotation (regarded as circular birefringence) *only in that the constant* $i\sigma_0$ *occurs in place of* ρ_0. Let $\partial''\vec{D}$ represent the partial increment to the initial vibration $\vec{D}$ due to the combined effects of optical rotation and circular dichroism (for the thickness dz). Then according to what has been said above $\partial''\vec{D}$ will be given by an expression which is similar to the expression for $\partial_3\vec{D}$ [which may be written down from equation (32)] except for the following difference: the constant ρ_0 will have to be replaced by $\bar{\rho}$ where

$$\bar{\rho} = \rho_0 + i\sigma_0. \tag{33}$$

In other words we will have

$$\left.\begin{aligned} \partial'' D_x &= -\frac{\pi}{\lambda_0} dz \cdot \bar{\rho} D_y \\ \partial'' D_y &= +\frac{\pi}{\lambda_0} \cdot dz \cdot \bar{\rho} D_x \end{aligned}\right\}. \tag{34}$$

If $\vec{D} + d\vec{D}$ represents the vibration at the plane $z + dz$ in the optically active absorbing crystal then $d\vec{D}$ represents the total increment to the initial vibration $\vec{D}$ under the combined effect of all the operations mentioned above. Thus $d\vec{D}$ is given by adding (31) and (34):

$$\left.\begin{aligned} dD_x &= -i\frac{2\pi}{\lambda_0}dz\,\{\bar{n}_{11}D_x + (\bar{n}_{12} - \tfrac{1}{2}i\bar{\rho})\,D_y\} \\ dD_y &= -i\frac{2\pi}{\lambda_0}dz\,\{(\bar{n}_{12} + \tfrac{1}{2}i\bar{\rho})\,D_x + n_{22}D_y\} \end{aligned}\right\}. \tag{35}$$

We wish to determine the *particular* states of polarisation of $\vec{D}$ for which the vibration is propagated unchanged with a specific complex velocity. From equation (14) we see that for such a disturbance we have

$$d\vec{D} = -i\frac{2\pi}{\lambda_0}dz\,.\,\bar{n}\vec{D} \tag{36}$$

Writing down the x and y components of (36) and comparing with (35) we finally obtain

$$\left.\begin{aligned} \bar{n} - \bar{n}_{11} &= (\bar{n}_{12} - \tfrac{1}{2}i\bar{\rho})\,\frac{D_y}{D_x} \\ \bar{n} - \bar{n}_{22} &= (\bar{n}_{12} + \tfrac{1}{2}i\bar{\rho})\,\frac{D_x}{D_y} \end{aligned}\right\} \tag{37}$$

which determine the characteristic states of polarisation D_y/D_x of the waves and their complex refractive indices $\bar{n}$—according to the method of superposition.

§ 8. Simplification of the Results of the Electromagnetic Theory

The equations (37) deduced by the superposition method are formally similar to the equations (14) deduced by the electromagnetic theory. It may be shown that the quadratic equation for D_y/D_x which may be obtained from (37) will have coefficients proportional to the corresponding coefficients in equation (16)—the factor of proportionality being $(-c/2v_m{}^3)$. It follows that the states of polarisation of the waves as obtained by the electromagnetic theory are *identical* with those obtained by the superposition method. Hence the points representing these states of polarisation on the Poincaré sphere‡ may be determined exactly as described in ref. 2—the parameters (ϕ, ψ) which specify the states of polarisation being the same in both methods.

‡ The Poincaré sphere merely represents a method of mapping states of polarisation, and hence need not be used only in conjunction with methods of superposition.

In the second place, *because of the formal similarity between equations* (37) *and* (14), it is clear that for every equation derived by the method of superposition a corresponding equation obtainable from the electromagnetic theory may be written down by inspection—merely by replacing the symbols according to a scheme which transforms (37) to (14). In addition to the replacement scheme already used for this purpose in inactive absorbing crystals (ref. 6, § 8), we have only to add that the symbols ρ_0 and σ_0 have to be replaced $-\ 2\Gamma_z$ and $-\ 2\Gamma_z'$ respectively. Simple expressions for the refractive indices and absorption coefficients of the waves have been derived in ref. 2, § 9 *c*, by the superposition method (using the Poincaré sphere). The corresponding equations from the electromagnetic theory will be:

$$v^2 = \tfrac{1}{2}(a_1 + a_2) + \tfrac{1}{2}\sqrt{(a_1 - a_2)^2 + (2\Gamma_z)^2} \cdot \cos 2\phi. \tag{38}$$

$$\kappa \cdot \frac{2v^3}{c} = \tfrac{1}{2}(b_1 + b_2) + \tfrac{1}{2}\sqrt{(b_1 - b_2)^2 + (2\Gamma_z')^2} \cdot \cos 2\psi. \tag{39}$$

For directions of propagation not in the vicinity of an optic axis it can be shown (as in ref. 2, § 7) that the velocities and absorption coefficients of the waves may be determined from the index and absorption ellipsoids. It is only for directions in the vicinity of an optic axis that the complications caused by the ellipticity of the waves make their appearance; for such directions, since the birefringence is necessarily small, the difference between the above expressions and those given by the method of superposition will not in general be of any practical significance.

§ 9. The Singular Axes

Among the most remarkable of the properties of absorbing crystals—both inactive and active—is the possibility of the occurrence of so-called *singular axes*. A singular axis represents a direction for which the quadratic equation (16)—which determines the states of polarisation of the waves—has equal roots; the condition ($b^2 = 4ac$) under which this can obtain ensures at the same time the equality of the roots of the quadratic equation (15) which determines the velocities of the waves. Thus along a singular axis there is only *one* particular state of polarisation which can be propagated without change of form. In a previous paper[3] we have already discussed (with reference to the particular example of amethystine quartz) the following problems: the conditions under which it is possible for singular directions to exist in optically active crystals, the location of these axes, and the state of (elliptic) polarisation of the single wave that can be propagated along a singular axis. In the present paper, therefore, we confine ourselves to the following interesting query: what will happen when a plane wave incident along a singular direction is in a state of polarisation *other* than that which *alone* can be propagated unchanged along that axis? In the particular case when the incident vibration is in a state of polarisation *orthogonal* to that of the wave which can be propagated unchanged along the singular axis, it had been supposed by Voigt[13] (in the case of inactive crystals) that the incident light would be totally reflected away, the reflection being partial in practical cases. That this is far from being the case has been shown experimentally by the author both in the case of iolite[7] (an optically inactive crystal) and amethyst[3] (an optically active crystal). The experimental results in the former case were readily explained[6] in detail (ref. 6, § 6 *b*) by a direct application of the method of superposition—according to which the incident vibration would obviously be propagated with a progressive change in its state of polarisation. *The same treatment and results apply* mutatis mutandis *to optically active absorbing crystals also.* Furthermore, it had been argued from considerations of continuity that the results given by the method of superposition could not really be contradictory to those of the electromagnetic theory. But how exactly the propagation of an arbitrary vibration along a singular axis could be directly handled by the electromagnetic theory was not apparent to the author till the publication of a recent paper by Jones[5]—in which is contained a new method of solving the electromagnetic equations in any anisotropic medium (transparent or absorbing). We present the method in a slightly modified form suitable for our present use.

§ 10. THE WAVE EQUATION FOR A PLANE WAVE PROPAGATED WITH CHANGE OF POLARISATION

In this section we wish to write down a suitable equation to represent a plane wave in a homogeneous medium, the wave being in general propagated with a progressive change in its state of polarisation. Firstly we shall take the vibration $\vec{D}$ at any plane z to be varying as $e^{i\bar{\omega}t}$ so that

$$\frac{\partial \vec{D}}{\partial t} = i\frac{2\pi}{\lambda_0} \cdot c\vec{D}. \tag{40}$$

Here the displacement vector has been written as a two-dimensional vector $\vec{D}$ since it is entirely transversal to the wave according to (3). As for the dependance of $\vec{D}$ on z, the form of the equation to be assumed is immediately suggested by equations (31) and (35) of § 7. The vibration at the plane $z + dz$ may be taken to be a linear *vector* function of the vibration at the plane z (at the same instant), since it is *not* assumed that both are necessarily in the same state of polarisation. We may then write

$$\frac{\partial \vec{D}}{\partial z} = -i\frac{2\pi}{\lambda_0}[n']\vec{D} \tag{41}$$

where $[n']$ is a two-by-two matrix operator. This resembles exactly the equation (36) which is satisfied by a usual plane wave of the form (4), but for the following difference: the refractive index $\bar{n}$ has been replaced by a 'refractive index tensor' $[n']$ in order that the same equation may represent a general plane wave propagated in a homogeneous anisotropic medium. This procedure is essentially in the same spirit as that of replacing the real refractive index by a complex quantity in order that the form of the usual equation to a plane wave in a transparent isotropic medium may still be retained to represent what is really a damped wave in an absorbing isotropic medium.

From (40) and (41), we obtain by differentiating with respect to t and z respectively

$$\frac{\partial^2 \vec{D}}{\partial t^2} = c^2[n']^{-2}\frac{\partial^2 \vec{D}}{\partial z^2}. \tag{42}$$

The above equation represents the wave equation satisfied by the disturbance propagated with change of polarisation; it resembles the usual form of the wave equation to a plane disturbance except that in place of the square of the velocity we have the matrix operator $c^2[n']^{-2}$.

As a simple example of a plane disturbance propagated with progressive change of polarisation we may remark that when a plane wave which is linearly polarised at a suitable azimuth is incident normally on a quarter-wave plate the incident linear vibration goes through progressive stages of elliptic polarisation and then emerges circularly polarised. This example also illustrates why, in usual cases, we need not directly seek general solutions representing disturbances propagated with change of polarisation: as will be evident in the example quoted, such a general solution is obtained by superposing the two characteristic plane wave solutions of the usual form (4)—*provided two such distinct solutions exist.*

§ 11. Second Method of Solving the Electromagnetic Equations

Our task is now to determine the refractive index tensor $[n']$ or alternatively the tensor $c^2[n']^{-2}$. The result is sufficiently elegant to be stated straightaway. Just as in an isotropic medium we have $n^2 = \epsilon$, where ϵ is the dielectric constant, in the present case it will be shown that

$$[n']^2 = [\epsilon], \tag{43}$$

where $[\epsilon]$ is the two-by-two matrix connecting $\vec{D}$ and $\vec{E}$:

$$\vec{D} = [\epsilon]\,\vec{E}. \tag{44}$$

Here $\vec{E}$ represents the 'projection' of the electric vector on the wavefront, being thus defined by the x and y components of the actual electric vector.

To establish (44) we note that the Maxwell's equations (2) may be written as

$$\frac{\partial^2 \vec{D}}{\partial t^2} = c^2 \frac{\partial^2 \vec{E}}{\partial z^2}. \tag{45}$$

The properties of the medium can be expressed in the form

$$c^2\vec{E} = [A]\,\vec{D} \tag{46}$$

where [A] is a two-by-two matrix whose components may be written down from equation (13).

From (45) and (46) we have

$$\frac{\partial^2 \vec{D}}{\partial t^2} = [A] \frac{\partial^2 \vec{D}}{\partial z^2}. \tag{47}$$

Comparing with (42) we find that whatever be the state of polarisation of $\vec{D}$ a solution of the form (41) is possible with

$$c^2 [n']^{-2} = [A]. \tag{48}$$

This relation is equivalent to (43) as may be seen by comparing (44) and (46). Thus the refractive index matrix $[n']$ can be determined§ for any direction of propagation from the relation

$$[n'] = c\,[A]^{-\frac{1}{2}} \tag{49}$$

We may briefly refer to the connection between the present method of solving the electromagnetic equations and that adopted in § 5. Though the wave equation (42) in general describes a disturbance propagated with

§ Only the physically significant square root of $[A]^{-1}$ is to be taken. For this and other mathematical questions which arise in the representation by matrix methods *see* Jones.[5,12]

a progressive change of polarisation this is not always the case. It will obviously reduce to the customary wave equation (for a homogeneously polarised disturbance) for those *particular* states $\vec{D}$ which satisfy the relation

$$[A]\,\vec{D} = \bar{v}^2\vec{D} \tag{50}$$

the complex velocity of the wave being $\bar{v}$. It will now be seen that the equations (13), previously used in § 5, are really the components of this vector equation, and as is shown by the procedure adopted there—the above equation is *usually* satisfied for two states of $\vec{D}$ (the eigenvectors of the matrix A) with *two* corresponding values of v^2 (the eigenvalues of the matrix A). For these particular states of $\vec{D}$ the equation (41) must also reduce to the form (36), *i.e.*, in the terminology of matrix calculus, these states of $\vec{D}$ are also the eigenvectors of the refractive index tensor $[n']$, the corresponding eigenvalues being the complex refractive indices of the waves.

From what has been said above, it is clear that a singular axis represents a special direction for which the matrix A has only *one* eigenvector, and correspondingly only *one* eigenvalue. This does not *in principle* lead to any difficulty in determining the refractive index matrix $[n']$ from (49), though of course the peculiar properties of [A] mentioned above are also carried over to $[n']$. On the other hand, when we adopt the method of superposition the refractive index tensor $[n']$ is considered as the sum of four parts each of which (it is assumed) may be directly determined in a simple fashion from the four corresponding surfaces defining the optical properties of the media. Thus whether we adopt the superposition method or the electromagnetic theory, no special difficulty arises in determining the refractive index tensor $[n']$ for propagation along a so-called singular direction. Along a singular direction—as, indeed, along any other direction—we can have plane disturbances which are propagated with a progressive change of polarisation: the speciality about a singular direction is, however, that such a disturbance cannot in turn be regarded as the sum of two plane waves of constant polarisation—there being only *one* wave of the latter type.

The author owes a debt of gratitude to Prof. Sir C. V. Raman, at whose instance the initial experimental investigations on amethyst and iolite were undertaken by the author, and without whose encouragement the subsequent theoretical investigations on the properties of absorbing crystals would not have been possible.

§ 12. SUMMARY

The propagation of light along an arbitrary direction in an absorbing active crystal is solved by extending the treatment previously given[1] for transparent active crystals—the index tensor and the modified gyration tensor being replaced by corresponding tensors with complex components. The two waves are in general elliptically polarised, in states identical with those given by a method of superposition[2]; their velocities and absorption coefficients are likewise simple functions of the parameters which specify these states of polarisation on the Poincaré sphere.

Attention is drawn to the propagation along any singular direction (in active or inactive crystals) where only *one* homogeneously polarised plane wave solution is obtained—and not two. A more general theoretical approach[5] becomes necessary to establish—in agreement with experiment—that other solutions also exist, representing plane disturbances propagated with a progressive change of polarisation. For such a disturbance the wave equation satisfied by the displacement vector differs from the usual form only in that the square of the velocity has to be regarded as a tensor operator.

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APPENDIX

(*a*) *Derivation of the constitutive relation in the form* (6).—The relation between **D** and **E** for an optically active absorbing crystal may be written as

$$\mathbf{D} = (\epsilon)\,\mathbf{E} + \mathbf{P}_1 + \mathbf{P}_2. \tag{46}$$

The portion of the induced polarisation $\mathbf{P}_1$ contributes to the mean absorption and the linear dichroism, while $\mathbf{P}_2$ leads to optical rotatory power and circular dichroism. These are given by the expressions (*see* ref. 9):

$$\mathbf{P}_1 = -\,i\,(\epsilon')\,\mathbf{E};\quad \mathbf{P}_2 = i\,\bar{\mathbf{G}} \times \mathbf{E}. \tag{47}$$

For usual values of absorption and optical activity $\mathbf{P}_1$ and $\mathbf{P}_2$ are so small that terms of the second order in $\mathbf{P}_1$ and $\mathbf{P}_2$ may be ignored. Hence to this degree of approximation $\mathbf{P}_1$ and $\mathbf{P}_2$ may be expressed as functions of **D** by substituting in (47) an approximate value of **E** obtained from (46), *viz.*, $\mathbf{E} = (\epsilon^{-1})\,\mathbf{D}$. We then obtain

$$\begin{aligned}(\epsilon)\,\mathbf{E} &= \mathbf{D} - \mathbf{P}_1 - \mathbf{P}_2\\ &= \mathbf{D} + i\,(\epsilon'\epsilon^{-1})\,\mathbf{D} - i\,(\mathrm{R}\epsilon^{-1})\,\mathbf{D}\end{aligned}$$

where the operator $\bar{\mathbf{G}}\times$ is replaced by an antisymmetric matrix operator R (*see, e.g.*, ref. 8, eq. 85·2). We then obtain

$$\mathbf{E} = (\epsilon^{-1})\,\mathbf{D} + i\,(\epsilon^{-1}\epsilon'\epsilon^{-1})\,\mathbf{D} - i\,(\epsilon^{-1}\mathrm{R}\epsilon^{-1})\,\mathbf{D}. \tag{48}$$

If we choose axes of co-ordinates X, Y, Z along the principal axes of the dielectric tensor (ϵ) it can be shown by actual matrix multiplication that the operator $(\epsilon^{-1}\mathrm{R}\epsilon^{-1})$ is equal to the operator $1/c^2 \cdot \bar{\Gamma}\times$, where

$$\bar{\Gamma}_{\mathrm{X}} = \frac{c^2}{\epsilon_{\mathrm{Y}}\epsilon_{\mathrm{Z}}}\cdot\bar{\mathrm{G}}_{\mathrm{X}};\quad \bar{\Gamma}_{\mathrm{Y}} = \frac{c^2}{\epsilon_{\mathrm{Z}}\epsilon_{\mathrm{X}}}\cdot\bar{\mathrm{G}}_{\mathrm{Y}};\quad \bar{\Gamma}_{\mathrm{Z}} = \frac{c^2}{\epsilon_{\mathrm{X}}\epsilon_{\mathrm{Y}}}\cdot\bar{\mathrm{G}}_{\mathrm{Z}}. \tag{49}$$

Comparing (48) with (6) and (8) we see that the vector of optical activity $\bar{\Gamma}$ is given by the above equations, while the index and absorption tensors are given by

$$(a) = c^2\,(\epsilon^{-1});\quad (b) = c^2\,(\epsilon^{-1}\epsilon'\epsilon^{-1}). \tag{50}$$

It must be remembered that $\bar{\mathbf{G}} = (\bar{g})\,\mathbf{s}$, where $(\bar{g})$ is the complex gyration tensor and **s** the wave-normal. If we write this relation in full (*see* ref. 9) it follows from (49) that the relation between the components of the tensor of optical activity $(\bar{\gamma})$ and the components of the complex gyration tensor $(\bar{g})$ will be given by

$$\bar{\gamma}_{1m} = \frac{c^2}{\epsilon_Y \epsilon_Z} \cdot \bar{g}_{1m}; \quad \bar{\gamma}_{2m} = \frac{c^2}{\epsilon_Z \epsilon_X} \bar{g}_{2m}; \quad \bar{\gamma}_{3m} = \frac{c^2}{\epsilon_X \epsilon_Y} \cdot \bar{g}_{3m} \qquad (51)$$

$$(m = 1, \ 2, \ 3).$$

(*b*) *An error in Jones' paper.*—In a paper by Jones[5] dealing with propagation in anisotropic media, the constitutive relation between **D** and **E** for an optically active medium has been expressed in a form which has been thought to be equivalent to that given above [eqs. (46) and (47)] but is really quite different from it. In particular the term $\mathbf{P}_2$ in (47) has been assumed by Jones to be given by

$$\mathbf{P}_2 = \frac{c}{\omega} \{(\bar{g}) \nabla\} \times \mathbf{E} \qquad (52)$$

where ∇ is the gradient operator and $(\bar{g})$ the complex gyration tensor. That this relation is quite different from the customary form (47) may be easily seen in the particular case when we seek plane wave solution of the usual form (4) so that $\nabla = i\omega \, \bar{n}/c \, . \, \mathbf{s}$. The relation (52) then becomes

$$\mathbf{P}_2 = i\bar{n} \, (\mathbf{G} \times \mathbf{E}). \qquad (53)$$

This is of course vitally different from the expression given in (47) since according to (53) the refractive index $\bar{n}$ which is one of the unknowns to be determined will itself be involved in the constitutive relations; more generally the refractive index tensor (n') will itself be involved in the matrix [A] occurring in (43). Accordingly, the relations given by Jones expressing his 'N-matrix' in terms of the complex dielectric and gyration-tensors are in error.

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