

THEORY OF OPTICAL ACTIVITY OF CRYSTALS

I. General Ideas

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1. INTRODUCTION

THE phenomenon of rotation of the plane of polarisation of light traversing an isotropic optically active medium was first explained by Fresnel, near about 1820, by supposing that right and left-circularly polarised light propagate with different velocities in the medium. The electromagnetic theory of light propagation in both isotropic and birefringent optically active media was developed by various authors, in particular by Voigt (1899, 1903) and Drude (1900). This theory is phenomenological in nature and it explains how optical activity varies with direction in a crystal and how it modifies the birefringence exhibited by it. Theories based on the structure of molecules (and crystals) have been proposed by various authors. A review of the theories, particularly in relation to the optical rotation of molecules, has been given by Condon (1937). The theories are, in general, of two types. In the first type, the molecule (or crystal) is treated as a system of coupled oscillators and a study of such a system in the field of the incident light wave leads to the evaluation of the rotatory power. The "coupled-oscillator" theory was first given, simultaneously and independently, by Oseen (1915) and Born (1915). Born's method was applied to the cases of sodium chlorate and bromate by Hermann (1923) and to β -quartz by Hylleraas (1927). Although these authors found a fairly good agreement with experiment, the calculations are so lengthy and complicated that the application of the method requires a large amount of labour. Kuhn (1929, 1930) gave a simple picture of a coupled oscillator and showed how it led to the presence of optical activity. On this basis, he has also derived the form of a dispersion equation for optical rotation (ρ) analogous to the Drude equation for refractive index. The coupled oscillator theory has been formulated in terms of quantum mechanics by Rosenfeld (1928).

The other type of theory, which may be termed the "polarisability theory", attempts to make use of purely optical principles for evaluating the magnitude of the rotation. The atoms, or groups of atoms, of which the

molecule (or crystal) is composed, are polarised in the presence of the field of the light wave. On summing up the induced moments of all the atoms, it would be found that the moment is not in phase with the field of the light wave, which is phenomenologically equivalent to the medium possessing optical activity. Theories of this type have been given by Gray (1916), de Mallemann (1924, 1930) and Boys (1934). Kirkwood (1937) has shown that, by introducing some simplifying assumptions (such as would lead to the additivity law for refraction), the polarisability theory can be derived from the general quantum mechanical formulation of coupled oscillators. Using the simplified result, he also calculated the magnitude of ρ for secondary butyl alcohol and found that it agreed reasonably well with experiment. Kirkwood also showed that earlier authors like de Mallemann and Boys, who made calculations in a practical case, had taken only second and third order terms of his formulation. In this present paper, it is proposed to give a classical picture of the first order terms in the polarisability theory. However, unlike Kirkwood, who applied the theory only to a molecule, we shall develop it for a crystal. In the first part, the general ideas of the theory are presented and are applied to a hypothetical crystal having a spiral structure, which would correspond to an optically active uniaxial crystal. The succeeding parts will contain applications to actual crystals.

The physical basis of the theory is as follows. The different atoms in a molecule, or near neighbours in a crystal, are at finite distances from each other, which, though small, would lead to a variation in the phase of the forced vibration excited by the incident light wave from atom to atom. Consequently, if we determine the influence of the electric moments induced in the neighbouring atoms on any particular atom, there would be an induced electric field which would not be in phase with that of the incident light wave. Under proper conditions, *e.g.*, when the electric vector of the light wave is parallel to a principal axis of the crystal, it would be found that there is a component of the induced electric field at right angles to the incident electric vector, which is 90° out of phase with it. Phenomenologically, this is equivalent to the medium possessing optical activity. As will be seen from the mathematics given below, if one calculates the induced moments in various directions, taking into account the interaction of the neighbouring atoms with the appropriate phases, then it is possible to obtain the magnitude of the optical rotation in all directions of the crystal. The interesting result emerges that, if all the atoms are isotropic, then the optical rotation becomes zero. Anisotropy of the individual atoms (or groups) appears to be essential for optical activity to be present in the first order. However, it may be present as a second or third order effect, *viz.*, the influence of one atom on

another and that again on a third atom, even if all the atoms are isotropic; but obviously the order of magnitude of such effects would be small compared with the first order.

2. ELECTROMAGNETIC THEORY OF LIGHT-PROPAGATION IN OPTICALLY ACTIVE CRYSTALS

The theory is discussed in detail in several treatises, for instance by Sziivessy (1928) and Born (1933). We shall merely quote the results that are of interest for the later sections.

If the co-ordinate axes are taken along the principal axes of the crystal, then the components of the displacement \mathbf{D} are related to those of the field \mathbf{E} by the relations

$$D_x = \epsilon_x E_x + i(\mathbf{G} \times \mathbf{E})_x, \text{ etc.} \quad (1)$$

where $\epsilon_x, \epsilon_y, \epsilon_z$ are the principal dielectric constants and \mathbf{G} is a vector whose components are given by

$$G_x = g_{11}s_x + g_{12}s_y + g_{13}s_z, \text{ etc.} \quad (2)$$

\mathbf{s} being a unit vector in the direction of propagation of the light wave. If n_0' and n_0'' are the two refractive indices for this direction of propagation in the absence of optical activity (i.e., as calculated from $\epsilon_x, \epsilon_y, \epsilon_z$), then the two indices in the presence of optical rotation are given by the roots of the equation:

$$(n^2 - n_0'^2)(n^2 - n_0''^2) = G^2, \quad (3)$$

where $G = \mathbf{s} \cdot \mathbf{G}$

$$\begin{aligned} &= g_{11}s_x^2 + g_{22}s_y^2 + g_{33}s_z^2 + (g_{23} + g_{32})s_y s_z + (g_{31} + g_{13})s_z s_x \\ &\quad + (g_{12} + g_{21})s_x s_y. \end{aligned} \quad (4)$$

The tensor g_{ij} is called the "gyration tensor" and completely describes the optical activity of the crystal.

In the directions of the two optic axes, $n_0' = n_0'' = \bar{n}$ (say). Then,

$$n_l = \bar{n} + G/2\bar{n}, \quad n_r = \bar{n} - G/2\bar{n}$$

and the rotatory power* is

$$\rho = \pi G/\lambda_0 \bar{n}, \quad (5)$$

where λ_0 is the wavelength of the light *in vacuo*.

* For mathematical convenience, and so as to agree with the definitions in later portions, ρ is positive for a left-rotating crystal and has the same sign as G . This is contrary to the usual definition of rotatory power, which is taken to be positive for right rotation cf. also Sziivessy, 1928).

For an arbitrary direction of propagation, the two velocities \bar{n}_0' and \bar{n}_0'' are given by

$$\begin{aligned}\bar{n}_0'^2 &= \frac{1}{2} \{n_0'^2 + n_0''^2 \mp |\sqrt{(n_0'^2 - n_0''^2)^2 + 4G^2}|\} \\ \bar{n}_0''^2 &= \frac{1}{2} \{n_0'^2 + n_0''^2 \pm |\sqrt{(n_0'^2 - n_0''^2)^2 + 4G^2}|\},\end{aligned}$$

where the upper or lower sign is to be taken according as $n_0' \leq n_0''$. Thus the phase difference between the two elliptically polarised waves per unit length, Δ , is given by

$$\Delta^2 = \frac{\pi^2}{\lambda_0^2 \bar{n}^2} (n_0'^2 - n_0''^2) + \frac{4\pi^2 G^2}{\lambda_0^2 \bar{n}^2} = \delta^2 + (2\rho)^2, \quad (6)$$

where δ is the corresponding phase difference when optical activity is absent and ρ is the optical rotation in the direction concerned and is given by the same formula as (5), \bar{n} now standing for the mean of the two refractive indices.

To make ideas clearer, we may take the case of an optically active cubic crystal, for which $\epsilon_x = \epsilon_y = \epsilon_z = \epsilon$ and $g_{11} = g_{22} = g_{33} = g$ (say) and all other components g_{ij} are zero. For light propagated along Oz, we have

$$\begin{cases} D_x = \epsilon E_x - ig E_y \\ D_y = \epsilon E_y + ig E_x \end{cases} \quad (7)$$

Thus, if the light is polarised with its electric vector along Ox, then, there is in addition to a displacement D_x also a component D_y at right angles to the electric vector, which is however 90° out of phase with it. Thus, if this perpendicular component is calculated from the structure, then the rotatory power, ρ , becomes calculable.

3. PRINCIPLE OF THE CALCULATION FROM CRYSTAL STRUCTURE

We shall use throughout Heaviside units for electrical quantities. Consider a crystal having p atoms, designated by 1, 2, ..., p , per unit cell. In some cases, it may be more convenient to take a group of atoms (such as a radical) as a single entity; however, we shall use the term atom in the general discussion to mean both a single atom as well as a group of atoms. The subscript r (1 to p) will denote an atom in the unit cell, while the subscript s will denote any general atom, including those in the unit cell. Let a_{ij}^r be the polarisabilities of the different atoms with respect to the chosen co-ordinate system, which has its axes parallel to the principal axes of the optical ellipsoid of the crystal. Let x_r', y_r', z_r' be the co-ordinates of the r th atom and let the incident polarised wave (propagated along positive z -axis) be represented by $E_x \exp ik(ct - z')$, ($k = 2\pi/\lambda$), λ being the wavelength in the medium. Leaving out the time variable $\exp ikct$, the mean electric field at the atom r is $(E_x + P_x/3) \exp -ikz_r'$, taking into account the Lorenz-

Lorentz field due to the surrounding medium. P_x is the electric moment per unit volume.

We wish now to find the electric field induced at atom r due to its neighbours. For this, choose atom r as origin, so that the co-ordinates of an atom s with respect to this are $x_s = x_s' - x_r'$, etc. The components of the moment induced in atom s by the field of the light wave alone neglecting the interaction of neighbours, are

$$\begin{aligned}\mu_{xs}' &= a_{11}'(E_x + P_x/3) \exp - ikz_s \\ \mu_{ys}' &= a_{12}'(E_x + P_x/3) \exp - ikz_s \\ \mu_{zs}' &= a_{13}'(E_x + P_x/3) \exp - ikz_s\end{aligned}\quad (8)$$

These induced moments would produce a field F_r (F_{xr} , F_{yr} , F_{zr}) at atom r . These are given by the formulæ:

$$F_{xr} = \frac{1}{4\pi} \sum_s \left(\frac{3x_s^2 - R_s^2}{R_s^5} \mu_{xs}' + \frac{3x_s y_s}{R_s^5} \mu_{ys}' + \frac{3x_s z_s}{R_s^5} \mu_{zs}' \right), \quad (9)$$

etc., where R_s is the distance between atoms r and s and the summation is to be performed over all atoms s that are significant. It may be noted that we have taken the field at r to be of the same phase as the moment of atom s . This is so because $R_s \ll \lambda$, the ratio being of the order of 1/1000 and it can be shown that, for an oscillating dipole, the field in its immediate vicinity has the same phase as the oscillations (Hertz, 1889). According to Hertz, the phase retardation ϕ at a distance R_s from the dipole is

$$\phi = kR_s - \tan^{-1} kR_s/(1 - k^2 R_s^2), \quad k = 2\pi/\lambda_0. \quad (10)$$

This, in fact, leads to a negative ϕ (or an advance of phase) for $R_s \sim \lambda_0/8$, ϕ becoming zero again at about $R_s = \lambda/4$. In the immediate vicinity of the oscillator, Eq. (10) gives

$$\phi = kR_s [1 - (1 + k^2 R_s^2)] = -k^3 R_s^3.$$

Thus, the phase change ϕ is two orders of magnitude smaller than kR_s (the corresponding change due to the incident wave) and can therefore be neglected in comparison with the latter. Since the phases of the different atoms s would, in general, be different from that of r , F_{xr} , F_{yr} , F_{zr} can be split into the form

$$F_{xr} = F_{xr}' + iF_{xr}'' = (a' + ia'')(E_x + P_x/3), \text{ etc.}$$

Thus, the components of the induced moment in atom r , taking the interaction of neighbours also into account, are

$$\mu_{xr} = a_{11}' \left(E_x + \frac{P_x}{3} + F_{xr} \right) + a_{12}' F_{yr} + a_{13}' F_{zr}, \text{ etc.} \quad (11)$$

Summing over the p atoms in the unit cell and dividing by the volume V of the unit cell, we obtain the components of the polarisation \mathbf{P} as

$$P_x = \frac{1}{V} \sum_p \mu_{xp} = (E_x + P_x/3) (A_x' + iA_x'') \quad (12)$$

and similar expressions for P_y and P_z . If these are compared with Eqs. (1), remembering that $\mathbf{D} = \mathbf{E} + \mathbf{P}$, then the values of ϵ_x and of some of the components of the gyration tensor g_{ij} can be obtained.

Repeating the calculations for the electric vector along Oy and similarly for directions of propagation along Ox and Oy , all the components of the tensors ϵ and g can be evaluated.

If the calculations are made correctly, the form of equations (1) would be the same as those deduced from the crystal structure—for instance, there would only be an imaginary part in the right-hand side of the expressions for P_y and P_z when the electric vector is parallel to Ox . The exact correspondence of the results from the molecular theory with the phenomenological expressions will be an indication of the correctness of the bases of the theory.

4. CALCULATION OF THE ROTATORY POWER ALONG THE OPTIC AXIS OF A HYPOTHETICAL UNIAXIAL CRYSTAL

In order to illustrate the procedure to be followed in making the calculations, we shall consider a tetragonal crystal having a simple spiral structure. The structure may be described as follows (Fig. 1).

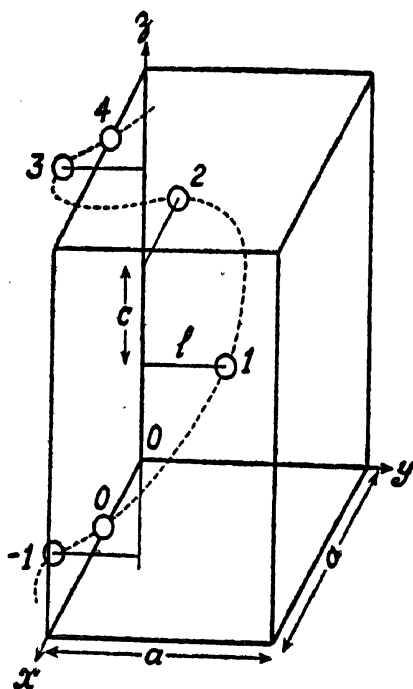


FIG. 1. Unit cell of hypothetical crystal

The unit cell has edges $a, a, 4c$. The c -axis is a four-fold screw axis (4_1), so that there is a spiral around Oz as axis, the atoms having the co-ordinates $l, 0, 0; 0, l, c; -l, 0, 2c; 0, -l, 3c$. Because of the translational symmetry of the lattice, there will be a similar spiral around each edge of the unit cell, parallel to Oz . The structure belongs to the space-group $D_4^3 - P4_12$, the other enantiomorphous form belonging to the space-group $D_4^7 - P4_32$. The crystal would thus exhibit point-group symmetry D_4 and is therefore uniaxial ($\epsilon_x = \epsilon_y \neq \epsilon_z$) and has only two independent components of the gyration tensor g_{11} and g_{33} ; $g_{22} = g_{11}$ and all other components are zero.

We suppose that l is small compared to a , say of the order of $\frac{1}{4}$, so that in calculating the interactions, the effect of atoms in one spiral on those in another may be neglected. Further, we shall only consider the influence of the two nearest neighbours in a particular spiral. The atoms are also supposed to be anisotropic, but for convenience, we take the principal axes of the polarisability ellipsoid to be along Ox, Oy , and Oz . Let the principal polarisabilities of the atom at level O ($z=0$) be a_1, a_2, a_3 along these directions. Then by reason of symmetry, those of atoms at levels 2 and 4 ($z=2c, 4c$) will be the same, while those of atoms 1 and 3 are a_2, a_1, a_3 respectively.

Consider a wave with electric vector along Ox travelling along Oz . Then the phase of an atom (x_r, y_r, z_r) is $\exp -ikz_r$ and the induced moments, neglecting the interaction of neighbours are

$$\begin{aligned} \mu_{rx}' &= a_1 (E_x + P_x/3) \exp -ikz_r & (r \text{ even}) \\ &= a_2 (E_x + P_x/3) & (r \text{ odd}) \end{aligned} \quad (13)$$

We thus have

$$\begin{aligned} F_{0x} &= \frac{1}{4\pi} \cdot \frac{(3l^2 - R^2)}{R^5} \cdot (\mu_{-1x} + \mu_{1x}) = \frac{1}{4\pi} \cdot \frac{(3l^2 - R^2)}{R^5} \cdot 2a_2 \cos kc (E_x + P_x/3) \\ F_{0y} &= \frac{1}{4\pi} \cdot \frac{3l^2}{R^5} \cdot (\mu_{-1x} - \mu_{1x}) = \frac{i}{4\pi} \cdot \frac{3l^2}{R^5} \cdot 2a_2 \sin kc (E_x + P_x/3) \end{aligned} \quad (14)$$

Similarly,

$$\begin{aligned} F_{1x} &= \frac{1}{4\pi} \cdot \frac{(3l^2 - R^2)}{R^5} \cdot 2a_1 \cos kc (E_x + P_x/3), \\ F_{1y} &= -\frac{i}{4\pi} \cdot \frac{3l^2}{R^5} \cdot 2a_1 \sin kc (E_x + P_x/3), \end{aligned} \quad (15)$$

$$F_{2x} = F_{0x}, F_{2y} = F_{0y}, F_{3x} = F_{1x}, F_{3y} = F_{1y} \quad (16)$$

Thus,

$$P_x = \frac{2}{V} \left[(a_1 + a_2) + \frac{1}{\pi} \cdot \frac{(3l^2 - R^2)}{R^5} \cdot a_1 a_2 \cos kc \right] (E_x + P_x/3) \quad (17)$$

$$P_y = \frac{i}{V} \cdot \frac{1}{\pi} \cdot \frac{3l^2}{R^5} (a_2^2 - a_1^2) \sin kc (E_x + P_x/3), \quad (18)$$

where V is the volume of the unit cell.

To take a numerical example, let $a = 6 \text{ \AA}$, $4c = 4 \text{ \AA}$, and let l the radius of the spiral be 1.5 \AA . The distance between nearest neighbours in the same spiral is 2.35 \AA while the nearest distance between atoms in two different spirals is 4.75 \AA , so that we can safely neglect the effect of the latter. Further, suppose $a_1 = 2.0$, $a_2 = 2.5$, $a_3 = 3.0 \times 10^{-23}$. The mean polarisability would roughly correspond to that of an oxygen atom. If we take the light to have a wavelength of 5000 \AA *in vacuo*, then $kc \sim 1/1000$, so that we can write $\cos kc = 1$ and $\sin kc = kc$. We then have

$$P_x = 0.664 (E_x + P_x/3); \quad P_y = i 5.94 \times 10^{-5} n (E_x + P_x/3).$$

From the former, we have $P_x/E_x = 0.853$, $n_\omega^2 = 1.853$, giving $n_\omega = 1.361$. Comparing the ratio of P_x to P_y with the phenomenological equations (1), we have

$$\frac{g_{33}}{n} = \frac{5.94 \times 10^{-5}}{0.664} (n^2 - 1) = 0.763 \times 10^{-4}$$

so that from (5), $\rho = 275^\circ$ per cm.

It is interesting [to note that the structure considered above gives a value for the optical rotation which is of the same order as is generally observed. For instance, quartz has almost the same rotation for light of this wavelength.

It may be noticed that P_x is real while P_y is pure imaginary as is required by the electromagnetic theory. The latter requires that, for the same direction of propagation, if the electric vector is along Oy , then P_y must have the same magnitude as P_x formerly, while P_x will have the same value as P_y before, but will be of the opposite sign. This can readily be verified.

5. ROTATORY POWER OF THE HYPOTHETICAL CRYSTAL FOR DIRECTIONS AT RIGHT ANGLES TO THE OPTIC AXIS

Consider light being propagated along positive Ox . If the electric vector is along Oy , the phenomenological equations are

$$D_y = \epsilon_y E_y = n_\omega^2 E_y; \quad D_z = i g_{11} E_y. \quad (19)$$

Calculations similar to those made in the previous section give

$$P_y = \frac{2}{V} \left[(a_1 + a_2) + \frac{1}{\pi} \cdot \frac{(3l^2 - R^2)}{R^5} \cdot a_1 a_2 \cos kl \right] \left(E_y + \frac{P_y}{3} \right) \quad (20)$$

$$P_z = - \frac{i}{V} \cdot \frac{1}{\pi} \cdot \frac{3lc}{R^5} a_3 (a_2 - a_1) \sin kl \left(E_y + \frac{P_y}{3} \right). \quad (21)$$

As before, $\cos kl = 1$, $\sin kl = kl$. Comparing (20) and (17) it is seen that the refractive index is the same as before, which is to be expected. Putting in numerical values,

$$P_z = -i \cdot 3 \cdot 96 \times 10^{-5} n (E_y + P_y/3),$$

which leads to a rotatory power $\rho_x = -183^\circ$ per cm. Thus the rotatory power at right angles to the optic axis is negative, *i.e.*, in the opposite sense to that along the optic axis, the ratio of the magnitudes being nearly $\frac{2}{3}$. This is not an unreasonable result, since quartz also exhibits a similar behaviour, the rotatory power at right angles to the optic axis being of the opposite sign to that along the optic axis, the ratio of the magnitudes of the two being $\frac{1}{2}$ (Bruhat and Grivet, 1935).

If the electric vector is along Oz, then we obtain

$$P_y = \frac{4}{V} \left[a_3 + \frac{1}{2\pi} \cdot \frac{(3c^2 - R^2)}{R^5} \cdot a_3^2 \cos kl \right] \left(E_z + \frac{P_z}{3} \right) \quad (22)$$

$$P_y = + \frac{i}{V} \cdot \frac{1}{\pi} \cdot \frac{3lc}{R^5} \cdot a_3 (a_2 - a_1) \sin kl \left(E_z + \frac{P_z}{3} \right). \quad (23)$$

The corresponding equations derived from Eq. (1) are

$$D_z = \epsilon_z E_z = n_e^2 E_z; \quad D_y = -ig_{11} E_z. \quad (24)$$

Comparing (23) and (21), it is seen that the coefficients on the right-hand side are the same, except for a change in sign as is to be expected from the second equations in (19) and (24). But from (22) we now obtain the extraordinary refractive index as $n_e = 1.378$. Thus although the refractive index is different, the rotatory power comes out to be the same, as should be the case if the calculations are correct.

Similarly, for light propagated along the Y-axis, detailed calculations bear out the deductions from Eq. (1), *viz.*,

$$D_x = n_\omega^2 E_x, \quad D_z = -ig_{11} E_x; \quad D_z = n_e^2 E_z, \quad D_x = ig_{11} E_z.$$

It is interesting to note that the magnitude of the rotation (both parallel and at right angles to the optic axis) is proportional to $(a_2 - a_1)$, so that they both change sign if $(a_2 - a_1)$ changes sign. Thus, the sense of optical rotation not only depends on the sense of the spiral, but also on the nature of the

anisotropy in the basal plane. In particular, if the atoms are completely isotropic (or at least isotropic in the xy -plane), the rotation vanishes. This is, however, an ideal case, for the very existence of a spiral structure would preclude such a possibility. The existence of neighbours arranged in a spiral would itself lead to anisotropy, just as the triangular group O_3 of three isotropic oxygen atoms would possess a strong anisotropy because of the arrangement (Bragg, 1924). The calculation of this anisotropy and the consequent rotations however, requires higher order terms.

6. SOME GENERAL CONSIDERATIONS

We have seen how, in the special case considered in the last two sections, the theory leads to results in conformity with the known results regarding optical rotation in a tetragonal crystal. It may be worthwhile to prove generally that the theory leads to the following two fundamental facts of optical activity.

(a) The rotation is of the same sense for opposite directions of propagation, and

(b) enantiomers will have opposite rotations.

To prove (a) we note that in Eqs. (8) and (9) all the quantities are unaltered except that $\exp -ikz_s$ is replaced by $\exp ikz_s$ when the direction of propagation is reversed. Thus, while the real parts of F_{xr} , F_{yr} , F_{zr} are unchanged, their imaginary parts will have opposite signs. But since the wave vector s in Eqs. (1) and (2) would also change sign, the components of the tensor g_{ij} would have the same sign irrespective of the sense of propagation.

To prove (b) we note that enantiomers are related to each other as figures inverted about a centre of inversion. Consequently the co-ordinates x_s , y_s , z_s of an atom s with respect to an atom r would *all* have opposite signs in the two cases. Thus, in Eq. (9), while terms like $3x_s^2 - R^2$, $3x_s y_s$ will have their signs unchanged, μ_s 's of the two enantiomers would be complex conjugates of each other because of the term $\exp -ikz_s$. Thus, in Eqs. (12) while the real parts are the same for both, the imaginary parts will be of opposite signs. Hence, the refractive indices of the two enantiomers will be the same, but the rotatory powers will be in opposite senses.

Although the theory is not intended to explain the dispersion of optical activity, it is interesting to note that it does explain it fairly accurately. Thus, it is found from the theory that, other conditions being the same, the rotatory power $\rho \propto 1/\lambda_0^2$. This comes from the factor $k (= 2\pi/\lambda)$ in the expression for g and the factor $1/\lambda$ in the expression for ρ in terms of g .

Since the polarisability α would also increase with decrease of wavelength, ρ would vary slightly faster than $1/\lambda^2$. This is in fact the case with a number of substances.

SUMMARY

The paper contains a classical description of the first order terms in the polarisability theory of optical activity. Optical activity essentially arises because the dipoles induced by the light wave in the medium are not all in the same phase. As a result of their mutual influences, the resultant induced moment will not be in phase with the electric field of the light wave and would lead to a rotation (ρ). By comparing the results calculated from the structure with the phenomenological theory of light propagation in the crystal, the magnitude of ρ is obtained. This method has been applied to a hypothetical tetragonal crystal having a spiral structure. It leads to the interesting result that the rotation at right angles to the optic axis is opposite in sign to that along the axis, as in quartz. The theory also predicts that, in general, the rotation will vary faster than $1/\lambda^2$, as is found to be the case with many substances.

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