

THE OPTICAL ROTATORY DISPERSION OF QUARTZ

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ABSTRACT

Many formulæ have been proposed to express numerically the rapid increase of the rotatory power of quartz with decrease of wavelength. An examination of these formulæ shows that they are wholly inappropriate for the case of quartz. It has been shown that the entire range of data from the visible to the extreme ultraviolet is accurately represented by a new type of formula involving only two constants, *viz.*,

$$\rho = \frac{k\lambda^2}{(\lambda^2 - \lambda_0^2)^2},$$

where $k = 7.19$ and $\lambda_0 = 0.0926283 \mu$. A theoretical interpretation of the new formula has been given on the basis of a simple coupled oscillator model.

INTRODUCTION

THE phenomenon of optical activity was discovered in the year 1811 by Arago in pieces of quartz cut perpendicular to the optic axis. Soon afterwards, Biot showed that the rotatory power in the visible region of the spectrum varies inversely as the square of the wavelength, *i.e.*, $\rho = k/\lambda^2$. The rotatory dispersion of quartz has subsequently been the subject of extensive experimental study. This has been reviewed by Sosman in his book on the properties of silica (1927) and by Lowry in his treatise on optical rotatory power (1935). More recently, measurements have been pursued into the remote ultraviolet upto 1500 Å by Servant (1939). It is evident from these determinations that Biot's Law fails completely as we move away from the visible spectrum. Indeed, from 6000 Å to 1500 Å the rotatory power increases by nearly 40 times instead of 16 times as required by the inverse square formula.

There have been several attempts in the past at expressing the rotatory dispersion in terms of assumed characteristic frequencies. In the present

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paper, these formulæ have been examined and it is shown that they are completely unsatisfactory for the case of quartz. A new type of formula has been suggested, a single term of which fits the whole range of data from the visible to the extreme ultraviolet very accurately. On the basis of a simple coupled oscillator model, a theoretical interpretation has been given to this formula. It is proved that the new formula is not open to the same theoretical objection which disqualifies all the earlier formulæ, which are principally of the Drude type.

EARLIER FORMULÆ FOR THE ROTATORY DISPERSION OF QUARTZ

The first attempt at a theoretical formula for the rotatory dispersion of quartz was made by Drude in his *Lehrbuch der Optik* (1900), where he proposed the form

$$\rho = \sum_r \frac{Q_r}{\lambda^2 - \lambda_r^2}, \quad (1)$$

where ρ is the rotatory power in degrees per millimetre, Q_r is a constant corresponding to the characteristic absorption wavelength λ_r in microns, the summation being performed over all the r absorption frequencies. Drude found that the then available data could be fitted by a two term formula, where

$$\begin{aligned} Q_1 &= 12.2, & Q_2 &= -5.046, \\ \lambda_1^2 &= 0.010627, & \lambda_2^2 &= 0, \end{aligned}$$

the negative term being unambiguously indicated to be necessary.

Lowry (1912) found that Drude's formula is not accurate enough to fit his first series of extended measurements and hence altered it to

$$\rho = \frac{11.6064}{\lambda^2 - 0.010627} - \frac{4.3685}{\lambda^2} + \frac{13.42}{\lambda^2 - 78.22}, \quad (2)$$

the additional third term corresponding to the infrared absorption band. The later and more accurate readings of Lowry and Coode-Adams (1927) required a further modification of (2), and their final formula is

$$\rho = \frac{9.5639}{\lambda^2 - 0.0127493} - \frac{2.3113}{\lambda^2 - 0.000974} - 0.1905, \quad (3)$$

where, instead of the single absorption band at 1031 Å used in the previous formulæ, two new characteristic wavelengths at 1129 Å and 312 Å have been used, the effect of the infrared being substituted by a constant.

However, all these formulæ fail to represent the measurements of Duclaux and Jeantet (1926) extending up to 1854 Å. These authors have proposed the formula

$$\rho = \frac{7.284}{\lambda^2 - 0.015114} \quad (4)$$

to approximately fit their data. But, as the discrepancy with this simple formula is quite considerable even in the near ultraviolet, another was worked out by Bradshaw and Livens (1929), viz.,

$$\begin{aligned} \rho = & \frac{845.694}{(\lambda^2 - 0.01274912)} - \frac{0.40235}{(\lambda^2 - 0.01274912)^2} \\ & - \frac{838.4320}{(\lambda^2 - 0.0120800)} - \frac{0.1331233}{(\lambda^2 - 0.0120800)^2} \\ & + \frac{43.05794}{(\lambda^2 - 80.00)} + \frac{2119.117}{(\lambda^2 - 80.00)^2} \end{aligned} \quad (5)$$

It will be noticed that the rotatory power in this formula is expressed as a difference of two very large terms of nearly equal magnitude but of opposite signs involving two absorption wavelengths respectively at 1129 Å and 1099 Å and that the influence of the infrared has been retained. The formula, however, deviates notably from the newer measurements of Servant (*loc. cit.*) in the far ultraviolet region. Servant (1940) has therefore fitted his results with the expression

$$\rho = 10^4 \frac{10^{-6}\lambda^2 + 0.515}{(13.956 \times 10^{-2}\lambda^4 - 17639 \times 10^{-6}\lambda^2 + 2.076)} \quad (6)$$

where λ is the wavelength of light in vacuum expressed in Å. He has assumed the existence of two characteristic wavelengths, one at 1064 Å, and another at 362 Å. The formula fits the entire data from 7000 Å to 1500 Å well. Later, the same author (1941), realising that a multiterm formula involving numerous unknown constants is very artificial and arbitrary, has rejected (6) in favour of the simpler formula

$$\rho = (n_0^2 + 2) \frac{165.6}{10^{-6}\lambda^2 - 1.325}, \quad (7)$$

where n_0 is the ordinary refractive index of quartz. The characteristic absorption assumed here lies at 1150 Å. The formula is approximately valid from 8000 Å to 2000 Å and fails farther out in the ultraviolet.

Finally, Radhakrishnan (1947) has proposed the formula

$$\rho = \frac{4.948}{\lambda^2 - 0.014161} + \frac{4.617}{\lambda^2 - 0.011236} - \frac{2.311}{\lambda^2 - 0.000974} - \frac{0.000815}{(\lambda^2 - 0.0195)^2} - 0.1905. \quad (8)$$

This one is largely a refinement on the work of Lowry and Coode-Adams. The absorption at 312 Å is still retained, but the single term at 1129 Å has been split into two terms at 1060 Å and 1190 Å. In addition a square term at about 1400 Å, almost at the limit of continuous absorption of light in quartz, has been introduced. The formula gives a tolerable fit.

Thus we see that a variety of formulæ involving numerous hypothetical absorption frequencies have been suggested to express the rotatory dispersion of quartz. Even a cursory examination of these formulæ shows that most of them are extremely complicated and arbitrary in character and all, except one, are unsatisfactory in accounting for the actual observed facts. Apart from this, there is an even more serious objection which may be raised against them.

KUHN'S SUMMATION RULE

A fundamental theorem concerning the optical rotatory power of molecular systems was derived by Kuhn (1929) in the first instance by considering the behaviour of a model consisting of two coupled resonators. The theorem states that the sum of the numerators determining the contributions of the different absorption frequencies to the rotatory power should vanish, *i.e.*,

if

$$\rho = \sum_r \frac{a_r \nu^2}{\nu_r^2 - \nu^2}, \quad (9)$$

then

$$\sum_r a_r = 0.$$

Proofs of this summation rule involving no special assumptions regarding the molecular model have since been given by several authors (*vide* reviews by Condon, 1937; Kauzmann, Walter and Eyring, 1940; Mathieu, 1946).

It is clear that the criterion furnished by this summation rule should be taken into account in deriving any formula, theoretical or empirical, which

claims to express rotatory dispersion. Without entering into the detailed numerical calculations, even a rough estimate of

$$\sum_r a_r (= \sum_r Q_r / \lambda_r^2)$$

in formulæ (1)–(8) shows that the summation rule is far from fulfilled by any one of them. As there is little justification in working out complicated expressions if we do not restrict ourselves to the basic requirement of the theory we may conclude that all the earlier formulæ are wholly inappropriate for the case of quartz.

A NEW FORMULA

The present writer has found (1952, 1954 *b*) that the rotatory dispersion of quartz from the visible to the extreme ultraviolet region of the spectrum is accurately represented by a simple formula involving only two constants, viz.,

$$\rho = \frac{k\lambda^2}{(\lambda^2 - \lambda_0^2)^2} \quad (10)$$

The formula approximates to Biot's Law when $\lambda \gg \lambda_0$. Also for a given value of λ_0 , the rate of variation of the rotatory power with wavelength in this formula is much greater than in the Drude type of formula. The constants which the experimental data are

$$k = 7.19, \quad \lambda_0^2 = 0.00858 \quad \text{or} \quad \lambda_0 = 0.0926283 \mu.$$

Here ρ is in degrees per millimetre and λ is in microns. The correct procedure is to express λ as the wavelength of light in vacuum. Servant has done so for his measurements in the remote ultraviolet, but the other authors have given the wavelengths in air. However, in the process of trying to fit a rotatory dispersion formula, Servant has effected the necessary corrections for a few wavelengths, extending from 1525 Å to 7000 Å. The rotatory powers for these wavelengths along with the values calculated with the above formula are set out below (Table I).

The agreement is very good from 6000 Å to 1525 Å. The error in this range, which never exceeds about two parts in a thousand, is *not of a progressive nature*. Data are available for many more wavelengths. The present writer (1954 *c*) has verified the validity of the formula for over 80 wavelengths extending from 6000 Å to 1520 Å. The fit is found to be extremely good over this range, the error being sometimes positive, sometimes negative. There is a slight discrepancy as we proceed towards the

TABLE I

$\lambda_{\text{vac.}}$ (in μ)	ρ (expt.)	ρ (calc.)	ρ (expt.)— ρ (calc.)
·1525	776·0	776·3	—0·3
·1550	724·0	724·1	—0·1
·1650	564·5	563·1	+1·4
·1750	453·5	453·1	+0·4
·1850	374·0	374·2	—0·2
·1950	315·5	315·3	+0·2
·2000	291·3	291·3	$\pm 0\cdot 0$
·2250	205·9	205·9	$\pm 0\cdot 0$
·2500	154·60	154·56	+0·04
·3000	97·72	97·61	+0·11
·4000	50·26	50·17	+0·09
·5000	30·86	30·84	+0·02
·6000	20·93	20·96	—0·03
·7000	15·13	15·20	—0·07

infrared from the red end of the spectrum. This is no doubt due to the contribution of the infrared bands which have not been included in the above formula. Lowry's measurements upto $3\cdot 7\ \mu$ definitely indicate the necessity of introducing a small infrared term.

It can easily be shown that the single characteristic wavelength at $0\cdot 0926283\ \mu$ used in the above formula lies in the region of the spectrum in which the ultraviolet bands of quartz appear. A one-term dispersion formula for the ordinary refractive index of quartz has been fitted up using this same wavelength. The formula proposed is of the Sellmeier-Drude type which is theoretically valid irrespective of whether a polarisation field exists or not (Krishnan and Roy, 1956). The formula is

$$n_{\omega}^2 - 1 = \frac{1\cdot 35\ \lambda^2}{\lambda^2 - \lambda_0^2}, \quad (11)$$

where

$$\lambda_0^2 = 0.00858.$$

λ (in μ)	n_w (expt.)	n_w (calc.)
·18547	1·676	1·673
·20006	1·649	1·649
·21107	1·634	1·635
·2428	1·605	1·606
·3034	1·577	1·578
·3587	1·564	1·564
·4800	1·550	1·550
·5893	1·544	1·544
·7065	1·540	1·540
·8447	1·538	1·538

The agreement is reasonably good over the whole range. This indicates that the characteristic wavelength at 0.0926283μ is approximately the effective average of the absorption spectrum of quartz in the ultraviolet. Of course, it is well known that the average characteristic wavelength operative in refractive dispersion and in rotatory dispersion formulæ need not necessarily be the same, simply because the form of the expressions in the two cases are different (Condon, *loc. cit.*), but in the particular case of quartz they happen to roughly coincide.

The rotatory power normal to the optic axis was measured for the first time by Voigt (1905). There have been a number of subsequent measurements, the most recent being those of Bruhat and Weil (1936), who have summarised their results as follows: the ratio of the rotatory powers of quartz perpendicular and parallel to the optic axis is independent of the wavelength for the spectral interval $5780 \text{ \AA} - 240 \text{ \AA}$, the ratio being -0.54 to within an accuracy of 0.5% .

Evidently, the rotatory dispersion normal to the optic axis should also be expressible by a formula of the same type as (10) with λ_0 unaltered and k reduced in the ratio of -0.54 .

DISCUSSION

In their article in the *Handbuch der Physik*, Born and Goppert-Mayer (1933) arrive at the general formula

$$\rho = \sum_r \frac{P_r \lambda^2}{(\lambda^2 - \lambda_r^2)^2} + \frac{Q_r}{(\lambda^2 - \lambda_r^2)} + \sum_v \frac{P_v \lambda^2}{(\lambda^2 - \lambda_v^2)^2} + \frac{Q_v}{(\lambda^2 - \lambda_v^2)} \quad (12)$$

It will be noticed that in addition to terms of the Drude type certain quadratic terms of the same form as suggested earlier are also present. We have now discovered that these terms play a predominant role in determining the rotatory dispersion of quartz. The simplicity of the new formula and the accuracy of its fit clearly shows that terms of the Drude type do not contribute in any observable measure to the rotatory power of quartz in the visible and ultraviolet region of the spectrum.

After this work had been completed, it was found on going through some of the very early literature on the subject that in 1883 Lommel had suggested a formula of the type

$$\rho = \frac{a\lambda^2}{(\lambda^2 - \lambda_0^2)^2}$$

for quartz and a few optically active liquids. He found that the available data for quartz from 8000 Å to 2200 Å were expressed tolerably well when $\log a = 0.8555912$ or $a = 7.17119$ and $\log \lambda_0^2 = 7.9341257 - 10$ or $\lambda_0 = 0.0928032 \mu$.

Lommel's work has apparently been overlooked by most of the authors who have since been interested in this crystal. We have now found that this formula fits the data even up to the extreme ultraviolet. In addition, as we shall see later, we have been able to give it a theoretical basis while actually Lommel's work was done before any proper theory of optical activity had yet been put forward.

THE THEORY OF COUPLED OSCILLATORS

The idea that optical activity arises from a particular spatial distribution of coupled oscillators was developed by Born, Oseen, Gray, de Malleman and others (*vide* reviews by Condon, *loc. cit.*; Kauzmann, *et al.*, *loc. cit.*; Mathieu, *loc. cit.*). Later, Kuhn contributed greatly to the subject by putting forward a simple model which demonstrated all the essential features of the phenomenon. Kuhn's model consists of two linear anisotropic resonators having natural frequencies of free vibration ν_1 and ν_2 respectively which are coupled with each other. Each resonator

is a charged particle which is assumed to be bound elastically to its own equilibrium position and capable of vibrating along a line. The analysis shows that the system has two modes of vibration, in each of which both resonators take part, one with frequency ν_1 and the other with frequency ν_2 . The contributions of the two modes to the rotatory power turn out to be of opposite signs when the frequency of the incident light is less than that of either resonator.

In this model ν_1 and ν_2 are assumed to be very different, so that the change of frequency brought about by the mutual coupling has been neglected altogether. This is, however, not permissible when ν_1 and ν_2 are very close to each other. In fact, in the limiting case when $\nu_1 = \nu_2$, each frequency is actually split into a doublet, the magnitude of the splitting being dependent on the strength of the interaction between the resonators.

We shall now discuss the optical behaviour of two identical linear anisotropic resonators coupled with each other. Let the natural frequency of vibration of the charged particle in each resonator be ν_0 , when uncoupled. As a result of the coupling, ν_0 would be split into two frequencies ν_1 and ν_2 . In one normal mode of vibration, the particles in the two resonators would vibrate with equal amplitude and in the same phase, and in the other normal mode they would vibrate with equal amplitude but opposite in phase.

Let the first resonator be situated at the origin of the co-ordinate system and let its vibration direction be OX. Let the second resonator be at $(0, 0, d)$ with its vibration direction parallel to OY. Suppose the charge and mass of the particle in each resonator to be q and M respectively. For convenience, we shall put $q = fe$ and $M = fm$, where e and m are the charge and mass of the electron and f the oscillator strength. We shall call this coupled system the *compound* resonator. Let us examine the action of a light wave on such a compound resonator.

Let right circular light given by

$$x = E_0 \cos \omega t$$

$$y = -E_0 \sin \omega t$$

be incident along OZ. The force exerted by the light wave on the charged particle of the first resonator is

$$F_1 = fe E_0 \cos \omega t.$$

The force on the charged particle of the second resonator which is rotated through 90° about the Z axis with respect to the first one is

$$F_2 = -fe E_0 \sin (\omega t - \phi),$$

where

$$\phi = \frac{2\pi\bar{n}d}{\lambda},$$

\bar{n} being the mean refractive index of the medium. Since ϕ is small

$$F_2 = -fe E_0 (\sin \omega t - \phi \cos \omega t).$$

Now, the compound resonator has two normal modes of vibration and the force may be expressed in terms of the two normal co-ordinates as

$$R_{\xi_1} = \frac{1}{\sqrt{2}} (F_1 + F_2)$$

$$R_{\xi_2} = \frac{1}{\sqrt{2}} (F_1 - F_2)$$

Substituting for F_1 and F_2 , we get

$$\begin{aligned} R_{\xi_1} &= \frac{fe E_0}{\sqrt{2}} [(1 + \phi) \cos \omega t - \sin \omega t] \\ &= a^{r_{\xi_1}} fe E_0 \cos (\omega t + \sigma), \end{aligned}$$

where $(a^{r_{\xi_1}})^2 = (1 + \phi)$, neglecting higher powers of ϕ . Hence, the equation of motion of the particles would be of the form

$$\ddot{\xi}_1 + \omega_1^2 \xi_1 = a^{r_{\xi_1}} \frac{fe E_0}{M} \cos (\omega t + \sigma).$$

Therefore,

$$\xi_1 = \frac{a^{r_{\xi_1}} fe E_0 \cos (\omega t + \sigma)}{M (\omega_1^2 - \omega^2)}$$

Hence, the dipole moment induced

$$\begin{aligned} &= (a^{r_{\xi_1}} fe)^2 \frac{E_0}{M (\omega_1^2 - \omega^2)} \\ &= (a^{r_{\xi_1}})^2 f \frac{e^2}{m} \frac{E_0}{\omega_1^2 - \omega^2} \end{aligned}$$

putting $M = fm$. The dipole moment could be expressed as two components at right angles to each other, say m_g and m_k , where

$$m_g = f \frac{e^2}{m} \frac{E_0}{\omega_1^2 - \omega^2}.$$

and

$$m_k = \phi f \frac{e^2}{m} \frac{E_0}{\omega_1^2 - \omega^2}.$$

These results are for a single compound resonator. If there are N such compound resonators all lying with their axes parallel to OZ but assuming different orientations about it, the mean polarisability per unit volume would be $N(m_g + m_k)/2 E_0$.

Hence

$$(n_r^2 - 1)_{\xi_1} = 2\pi N (a^r_{\xi_1})^2 \frac{e^2}{m} \frac{f}{\omega_1^2 - \omega^2}.$$

In a similar manner, we get for the other normal mode

$$(n_r^2 - 1)_{\xi_2} = 2\pi N (a^r_{\xi_2})^2 \frac{e^2}{m} \frac{f}{\omega_2^2 - \omega^2},$$

where

$$(a^r_{\xi_2})^2 = 1 - \phi.$$

Let us write

$$n_r^2 - 1 = (n_r^2 - 1)_{\xi_1} + (n_r^2 - 1)_{\xi_2}.$$

For left circular light given by

$$x = E_0 \cos \omega t$$

$$y = E_0 \sin \omega t,$$

we have similarly

$$(n_l^2 - 1)_{\xi_1} = 2\pi N (a^l_{\xi_1})^2 \frac{e^2}{m} \frac{f}{\omega_1^2 - \omega^2},$$

$$(n_l^2 - 1)_{\xi_2} = 2\pi N (a^l_{\xi_2})^2 \frac{e^2}{m} \frac{f}{\omega_2^2 - \omega^2},$$

where

$$(a^l_{\xi_1})^2 = 1 - \phi,$$

$$(a^l_{\xi_2})^2 = 1 + \phi,$$

and

$$n_l^2 - 1 = (n_l^2 - 1)_{\xi_1} + (n_l^2 - 1)_{\xi_2}.$$

Therefore,

$$n_l^2 - n_r^2 = 4\pi N \frac{e^2}{m} f \phi \left[\frac{1}{\omega_2^2 - \omega^2} - \frac{1}{\omega_1^2 - \omega^2} \right].$$

It will be noticed that the numerators in the two terms are equal in magnitude but opposite in sign and this is in conformity with Kuhn's summation rule.

Let

$$\omega_1^2 = \omega_0^2 + 2\pi^2\epsilon$$

$$\omega_2^2 = \omega_0^2 - 2\pi^2\epsilon$$

We shall assume that the resonators are not very strongly coupled so that ϵ is a small quantity. Putting

$$n_l + n_r = 2\bar{n}, \quad \phi = \frac{2\pi\bar{n}d}{\lambda}$$

and

$$\omega_0 = 2\pi\nu_0,$$

we get

$$n_l - n_r = \frac{Ne^2d}{m\lambda} \frac{f\epsilon}{(\nu_0^2 - \nu^2)^2}$$

neglecting ϵ^2 in the denominator. Hence

$$\rho = \frac{\pi}{\lambda} (n_l - n_r) = \frac{A_0\lambda^2}{(\lambda^2 - \lambda_0^2)^2},$$

where

$$A_0 = \frac{\pi Ne^2d\lambda_0^4 f\epsilon}{mc^4}.$$

If the compound resonators were randomly oriented we would have to introduce a factor $\frac{1}{3}$ in the expression for the rotatory power.

If, as a more general case, we assume that the first resonator $(0, 0, 0)$ has direction cosines α, β, γ and that the second resonator $(0, 0, d)$ is rotated through an angle θ about the Z axis with respect to the first one, it may be shown that

$$\rho = \frac{A_0\lambda^2}{(\lambda^2 - \lambda_0^2)^2}, \quad (13)$$

where

$$A_0 = \frac{\pi Ne^2d \sin \theta (\alpha^2 + \beta^2) \lambda_0^4 f\epsilon}{3 mc^4}$$

It will be seen from (13) that the two enantiomorphous forms will have opposite rotatory powers, since $\rho \propto \sin \theta$.

Thus, by means of an extremely elementary model we have been able to derive the correct expression for the rotatory dispersion of quartz. It should be emphasised that the new single term formula intrinsically satisfies Kuhn's summation rule and is therefore more justified than any of the earlier formulæ worked out for this crystal.

We shall now try and interpret these results in terms of the structure of quartz. α -Quartz belongs to the space group D_3^4 or the enantiomorphous D_3^6 . The unit cell has three SiO_2 units spaced at equal intervals along the vertical screw axis, each of which is turned through 120° with respect to its predecessor. It is this screw structure which is responsible for its optical activity, the individual constituent units themselves being optically inactive. In view of the foregoing theoretical considerations, it can scarcely be doubted that the rotatory power of quartz arises primarily as a result of an interaction in the nature of a resonance between the similar polarisable units constituting the crystal. This type of interaction causes a splitting of the characteristic frequency of each individual unit and consequently, the rotatory dispersion formula is of the form (13). By a simple extension of the model which we have just considered, it may be shown in an approximate way (Chandrasekhar, 1954 *c*) that for an hypothetical crystal with a screw structure built up of such compound resonators, the rotatory dispersion formula for propagation of light along and normal to the optic axis are both of the quadratic form (13), a result which agrees with the experiments of Bruhat and Weil (*loc. cit.*).

The same type of formula is also found to fit the data for cinnabar, benzil and sodium chlorate (Chandrasekhar, 1953, 1954 *a*, 1954 *c*) quite well and, in fact, much better than any of the previous formulæ proposed for these crystals. It may be remarked that, like quartz, the rotatory power of these crystals is present only in the crystalline state and not in solution.

In conclusion, I have great pleasure in expressing my thanks to Professor Sir C. V. Raman, F.R.S., for the valuable discussions I had with him during the course of these investigations. I am also grateful to Dr. S. Ramaseshan for his kind advice and interest.

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