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THE OPTICAL ROTATORY POWER OF QUARTZ AND ITS VARIATION WITH TEMPERATURE

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1. INTRODUCTION

SOON after the discovery of the optical activity of quartz by Arago in 1811, Biot showed that the magnitude of the rotation in the visible region of the spectrum is inversely proportional to the square of the wavelength, *i.e.*, $\rho = k/\lambda^2$. The rotary dispersion of quartz has been, subsequently, the subject of numerous investigations. These have 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, the measurements have been extended into the ultra-violet upto 1525 A.U. by Servant (1938) and into the infra-red upto $9{\cdot}7\mu$ by Gutowsky (1951). From these determinations it is evident that Biot's Law fails completely as we move away from the visible region of the spectrum in either direction. Indeed, in the ultra-violet, the rotatory power increases from 6000 A.U. to 1500 A.U. nearly 40 times instead of 16 times as required by the inverse square formula. Several attempts have been made to represent the rotary dispersion numerically in terms of various assumed dispersion frequencies. A critical examination of these formulæ, however, shows that they are arbitrary in character and do not possess any real physical justification.

A noteworthy fact about the optical rotatory power of quartz is that it increases appreciably when the temperature of the crystal is raised, the rate of increase accelerating as the temperature approaches that at which the α-β transformation in crystalline quartz takes place. At the transition point itself, there is an abrupt increment in the rotatory power. The temperature-coefficient remains positive in β-quartz but is very much smaller than in α-quartz. Though many experimental studies dealing with these effects are on record, no attempt appears to have been made to give a theoretical explanation of the influence of temperature on the optical activity of quartz.

It is proposed to discuss in this paper, firstly, the manner in which the optical rotatory power of quartz varies with wave-length and secondly, its

variation with the temperature of the crystal. It emerges from the discussion that the two topics are closely interrelated.

Other crystals, *viz.*, cinnabar and sodium chlorate, are known which are optically active and in which also the temperature coefficient of the optical rotatory power is positive. These cases will be discussed in later papers.

2. THE EARLIER FORMULÆ FOR THE ROTARY DISPERSION

The first attempt at a theoretical formula to fit the rotary dispersion of quartz was made by Drude in his "Optics" where he proposed the form

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

where ρ is the optical 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

$$Q_1 = 12{\cdot}2 \qquad Q_2 = -5{\cdot}046$$
$$\lambda_1^2 = 0{\cdot}010627 \qquad \lambda_2^2 = 0.$$

the negative term being unambiguously indicated to be necessary.

Since then several other modified formulæ have been suggested by various authors. Lowry (1912) noticed that Drude's formula is not accurate enough to fit his first series of extended measurements and hence altered it to

$$\rho = \frac{11{\cdot}6064}{\lambda^2 - 0{\cdot}010627} - \frac{4{\cdot}3685}{\lambda^2} + \frac{13{\cdot}42}{\lambda^2 - 78{\cdot}22} \tag{2}$$

the additional third term corresponding to the infra-red absorption band. His later still more accurate readings upto 2263 A.U. however required still further modification and his final formula is

$$\rho = \frac{9{\cdot}5639}{\lambda^2 - 0{\cdot}0127493} - \frac{2{\cdot}3113}{\lambda^2 - 0{\cdot}000974} - 0{\cdot}1905 \tag{3}$$

where instead of the single absorption band at 1031 A.U. occurring in the previous formulæ, two new characteristic wave-lengths 1129 A.U. and 312 A.U. have been used, the effect of the infra-red being substituted by a constant.

Neither of these latter formulæ, however, represent the results of Duclaux and Jeantet (1926) which extend upto 1854 A.U. and therefore another formula was worked out by Bradshaw and Livens (1929), *viz.*,

$$\rho = \frac{845\cdot694}{(\lambda^2 - 0\cdot01274912)} - \frac{0\cdot40235}{(\lambda^2 - 0\cdot01274912)^2} - \frac{838\cdot4320}{(\lambda^2 - 0\cdot0120800)} - \frac{0\cdot1331233}{(\lambda^2 - 0\cdot0120800)^2} + \frac{43\cdot05794}{(\lambda^2 - 80\cdot00)} + \frac{2119\cdot117}{(\lambda^2 - 80\cdot00)^2}. \quad (4)$$

It will be noticed that in this formula, the optical rotatory power is expressed as a difference of two very large terms of nearly equal magnitude but of opposite signs involving two close absorption wave-lengths respectively at 1129 A.U. and 1099 A.U., and that the influence of the infra-red term has been retained. Though some additional terms of a different type occur in this formula, it is clear that the terms of the Drude type appearing in it are the principal ones. The formula, however, gives values which deviate notably from the newer extended measurements of Servant (*loc. cit.*) in the remote ultra-violet. Radhakrishnan (1947) has therefore fitted these results with still another rotary dispersion formula, *viz.*,

$$\rho = \frac{4\cdot948}{\lambda^2 - 0\cdot014161} + \frac{4\cdot617}{\lambda^2 - 0\cdot011236} - \frac{2\cdot311}{\lambda^2 - 0\cdot000974} - \frac{0\cdot000815}{(\lambda^2 - 0\cdot0195)^2} - 0\cdot1905 \quad (5)$$

This formula is largely a refinement on the work of Lowry and Coode Adams. The absorption at 312 A.U. is still retained but the single term at 1129 A.U. has been split into two terms at 1060 A.U. and 1190 A.U. In addition, a small negative term at about 1400 A.U., almost at the very limit of continuous absorption in quartz has been introduced. This formula fits the entire data fairly well.

Thus, we see that a variety of formulæ involving numerous hypothetical absorption frequencies have been suggested to numerically represent the rotary dispersion of quartz. On examining these formulæ, we notice that, without exception, all the authors have been obliged to introduce negative terms involving absorption wave-lengths lying in the ultra-violet, in order to fit the data over the whole spectrum. No physical justification has been given for this procedure. The necessity for assigning a negative strength to a term becomes clear when we consider a single-term formula of the type (1) as suggested by Drude. When $\lambda_r = 0$, this reduces to Biot's Law. When λ_r takes finite values, the rotatory power varies faster with wave-length than required by Biot's Law. The rotatory power has been calculated for several wave-lengths giving various values to λ_r in this formula and shown in

Table I. These values of λ_r have been chosen at random over the entire region of continuous absorption in quartz which extends below 1436 A.U. (Tousey, 1940). In each column, the rotatory power has been fitted with the experimental value for $\lambda = 0{\cdot}6708\,\mu$. (Throughout, ρ is expressed in degrees per millimetre.)

TABLE I

Rotary dispersion using one-term formula of Drude type

λ (*in* μ)	ρ (Calculated)					ρ (Experimental)
	$\lambda_r = 0$	$\lambda_r = \cdot 03\mu$	$\lambda_r = \cdot 06\mu$	$\lambda_r = \cdot 10\mu$	$\lambda_r = \cdot 12\mu$	
·1525	320	332	375·6	549	813	776
·1625	281·8	291·2	323·7	443·5	600·0	599·5
·1825	228·4	229·1	248·5	312·2	381·0	391·5
·2359	133·7	135·6	141·8	159·4	174·6	180·4
·3728	53·5	53·8	54·5	56·4	57·8	58·8
·4678	34·0	34·1	34·3	34·8	35·2	35·6
·6708	16·54	16·54	16·54	16·54	16·54	16·54

It is seen that for all values of λ_r, except $\lambda_r = 1200$ A.U., the calculated rotatory power is much lower than the observed value over the whole range of wave-lengths. The same is true even for $\lambda_r = 1200$ A.U. except in the remote ultra-violet region where the calculations exceed the observed data. On a careful study of this table it can easily be understood why no combination of positive terms can ever accurately represent the rotary dispersion of quartz. For, even if at the two extremes of the spectrum the values be accurately fitted by a suitable choice of terms, there will be a large region in the visible and near ultra-violet for which the calculated values will be less than the experimental values. This difficulty has been overcome by the previous authors by expressing the rotatory power as a difference of two terms. Evidently, these formulæ are purely empirical in character and do not give a real explanation of the facts. Hence, we may conclude that a rotary dispersion formula of the type suggested by Drude is not appropriate in the case of quartz.

3. A NEW FORMULA

The present author has found that the rotary dispersion of quartz is accurately represented throughout the visible and ultra-violet range of the spectrum by a simple formula containing only two constants, *viz.*,

$$\rho = \frac{k\lambda^2}{(\lambda^2 - \lambda_0^2)^2}. \qquad (6)$$

It is seen that the formula approximates to Biot's Law when $\lambda >> \lambda_0$. Also for a given value of λ_0, the rate of variation of the rotation with wave-length from this formula is much greater than from the Drude formula. The constants which fit the experimental data are

$$k = 7{\cdot}186 \text{ and } \lambda_0 = 0{\cdot}0926283\,\mu.$$

The rotatory power calculated from this formula are given in comparison with the experimental values in Table II.

TABLE II

Rotary dispersion using author's formula

λ (in μ)	ρ (experimental)	ρ (calculated)	λ (in μ)	ρ(experimental)	ρ (calculated)
·1525	776	776	·291216	104·97	104·88
·1575	724	724	·340365	72·46	72·35
·1625	599·5	597·2	·372762	58·84	58·75
·1675	533·0	531·5	·435834	41·55	41·50
·1700	504·5	503·0	·467816	35·61	35·57
·1750	453·5	452·8	·479991	33·68	33·65
·1775	431·5	430·7	·508582	29·73	29·72
·1800	410·5	410·3	·520908	28·25	28·24
·1825	391·5	391·5	·546074	25·54	25·55
·1854	370·9	371·3	·579066	22·55	22·57
·1935	322·76	322·89	·643847	18·02	18·08
·198979	295·65	295·82	·670785	16·54	16·60
·217402	226·91	226·96	·810	11·11	11·24
·226909	200·90	200·96	·940	8·14	8·29
·235923	180·43	180·44	1·170	5·18	5·32
·241331	169·68	169·70	1·342	3·89	4·03
·262830	135·66	135·62	2·500	0·98	1·15
·273955	122·12	122·06	3·210	0·52	0·70

While the previous formulæ, with the single exception of Radhakrishnan's fail in the remote ultra-violet, this one fits very well from 6000 A.U. to 1525 A.U. In fact from 5800 A.U. to 1800 A.U., the maximum error is only about one part in a thousand, being sometimes positive, sometimes negative. Between 1800 A.U. and 1525 A.U., the deviation is slightly larger for certain wavelengths, but even so it is not of a progressive nature. There is a small discrepancy as we proceed from the red end of the spectrum to the infra-red, the error uniformly increasing with increase of wave-length. This is no doubt due to the contribution of the infra-red absorption bands, which have not been included in the above formula. As a matter of fact, Gutowsky (*loc. cit.*) has found that quartz exhibits anomalous rotary dispersion beyond 3·7 μ, the rotation, instead of decreasing as is to be expected,

increasing nearly four times from $3{\cdot}7\,\mu$ to $9{\cdot}7\,\mu$, thereby indicating that the presence of absorption bands in the infra-red has an influence on the optical rotatory power.

It can easily be shown that the single characteristic frequency used in the above rotary dispersion formula lies within the region of the ultra-violet spectrum in which the absorption frequencies of quartz appear. A one-term dispersion formula for the ordinary refractive index has been fitted up using this same frequency. The formula proposed is of the Sellmeyer-Drude type which is known to yield satisfactory results in the case of quartz (Radhakrishnan, 1951). The formula is

$$n_w^2 - 1 = \frac{1{\cdot}35\,\lambda^2}{\lambda^2 - (0{\cdot}0926283)^2}. \tag{7}$$

The calculated values are given in Table III.

TABLE III

Dispersion of quartz (for explanation see text)

λ (in μ)	n (experimental)	n (calculated)
·1854	1·676	1·673
·2413	1·605	1·607
·3404	1·5675	1·5678
·5086	1·548	1·548
·6708	1·541	1·541
·8000	1·538	1·539
1·2000	1·532	1·536

We see that the error in the calculated value over the whole range is only in the third decimal place. This indicates that the characteristic wave-length of 926 A.U. is roughly the same as the effective average wave-length of the absorption spectrum of quartz in the ultra-violet.

4. THE TEMPERATURE VARIATION OF THE OPTICAL ROTATORY POWER

The rotary dispersion formula (6) may be expressed in the form

$$\rho = \frac{a\nu^2}{(\nu_0^2 - \nu^2)^2}, \tag{8}$$

where 'a' is a constant such that $k = a\,\lambda_0^4$ and ν_0 is the characteristic absorption expressed in wave-numbers. The optical rotatory power of quartz is known to increase with rise of temperature even when the expansion

along the optic axis has been corrected for. To calculate this increment let us differentiate equation (8) with respect to temperature. We shall simply consider 'a' to be constant with temperature, but ν_0 to vary with it, so that

$$\frac{d\rho}{dt} = \frac{4a\nu^2\nu_0^2}{(\nu_0^2 - \nu^2)^3}\chi_0, \tag{9}$$

where

$$\chi_0 = -\frac{1}{\nu_0}\frac{d\nu_0}{dt} = \frac{1}{\lambda_0}\frac{d\lambda_0}{dt}.$$

Hence the temperature coefficient of the optical rotatory power

$$\frac{1}{\rho}\frac{d\rho}{dt} = \frac{4\lambda^2}{(\lambda^2 - \lambda_0^2)}\chi_0. \tag{10}$$

Molby (1910) has made precision measurements of the optical rotatory power of quartz at temperatures of $-118°$ C. and 20° C. from 4000 A.U. to 6700 A.U. With the aid of equation (9) and the rotatory dispersion formula (6), the values of $d\rho/dt$ have been theoretically accounted for over the whole range of wave-lengths. The value of χ used is $2{\cdot}7 \times 10^{-5}$. The theoretical values are given in Table IV.

TABLE IV

Temperature variation of optical rotatory power

λ (in μ)	ρ (experimental)		$\frac{d\rho}{dt}$ (expt.)	$\frac{d\rho}{dt}$ (theor.) $\chi = 2{\cdot}7\times10^{-5}$
	20° C.	−188° C.		
·4047	48·842	47·675	$5{\cdot}6\times10^{-3}$	$5{\cdot}6\times10^{-3}$
·4359	41·441	40·469	$4{\cdot}7\times10^{-3}$	$4{\cdot}7\times10^{-3}$
·4670	35·529	34·696	$4{\cdot}0\times10^{-3}$	$4{\cdot}0\times10^{-3}$
·4800	33·593	32·818	$3{\cdot}7\times10^{-3}$	$3{\cdot}8\times10^{-3}$
·5086	29·656	28·983	$3{\cdot}2\times10^{-3}$	$3{\cdot}3\times10^{-3}$
·5461	25·472	24·885	$2{\cdot}8\times10^{-3}$	$2{\cdot}8\times10^{-3}$
·5893	21·686	21·188	$2{\cdot}4\times10^{-3}$	$2{\cdot}4\times10^{-3}$
·6438	18·003	17·603	$1{\cdot}9\times10^{-3}$	$2{\cdot}0\times10^{-3}$
·6708	16·493	16·145	$1{\cdot}7\times10^{-3}$	$1{\cdot}8\times10^{-3}$

The agreement between the theoretical and experimental values is satisfactory. From equation (10) we notice that the theory predicts that the temperature coefficient of the rotatory power should exhibit an increase with decrease of wave-lengths. This in fact is known to be true from Molby's measurements and also from the observations of Soret and Sarasin (1882).

Le Chatelier (1889) has made measurements of the rotatory power over wide ranges of temperatures both below and above the α-β transformation

point in quartz. Using a value of $\chi = 4{\cdot}5 \times 10^{-5}$, $d\rho/dt$ has been theoretically calculated as before. These are given in Table V.

TABLE V

Temperature variation of optical rotatory power

λ (in μ)	$\frac{d\rho}{dt}$ (experimental)	$\frac{d\rho}{dt}$ (theor.) $\chi = 4{\cdot}5 \times 10^{-5}$
·279	$27{\cdot}5 \times 10^{-3}$ (20° – 475° C.)	24×10^{-3}
·448	$7{\cdot}0 \times 10^{-3}$ (20° – 415° C.)	$7{\cdot}3 \times 10^{-3}$
·500	$5{\cdot}4 \times 10^{-3}$ (,,)	$5{\cdot}7 \times 10^{-3}$
·518	$5{\cdot}0 \times 10^{-3}$ (,,)	$5{\cdot}3 \times 10^{-3}$
·589	$4{\cdot}3 \times 10^{-3}$ (,,)	$4{\cdot}0 \times 10^{-3}$
·656	$3{\cdot}4 \times 10^{-3}$ (,,)	$3{\cdot}2 \times 10^{-3}$

The value of χ in this case has increased as the temperature is higher. In fact, as will be seen later, χ is found to have almost the same value and increase in roughly the same proportion with temperature, from the refraction data as well. Thus the increase of temperature coefficient with increase of temperature and with decrease of wave-length have both become intelligible from these calculations.

5. RELATION TO ORDINARY DISPERSION AND ITS THERMAL VARIATION

It would be of interest to see how the results obtained in the previous section are related to ordinary dispersion and its variation with temperature. Dispersion formulæ for quartz have been suggested by many authors in the past. We shall consider here only that put forward by Radhakrishnan (*loc. cit.*) which has the merit of fitting both the ordinary and extraordinary indices very accurately. We shall quote only that for the ordinary index, *viz.*,

$$n_w^2 - 1 = \frac{0{\cdot}663044\,\lambda^2}{\lambda^2 - (0{\cdot}0600)^2} + \frac{0{\cdot}517852\,\lambda^2}{\lambda^2 - (0{\cdot}1060)^2} + \frac{0{\cdot}175912\,\lambda^2}{\lambda^2 - (0{\cdot}1190)^2} + \frac{0{\cdot}565380\,\lambda^2}{\lambda^2 - (8{\cdot}844)^2} + \frac{1{\cdot}675299\,\lambda^2}{\lambda^2 - (20{\cdot}742)^2}. \qquad (11)$$

In this formula, the absorption at 600 A.U. is a hypothetical one, while those at 1060 A.U. and 1190 A.U. correspond with the reflection maxima observed by Tousey (*loc. cit.*). In addition, there are two small infra-red terms which serve to give a very accurate fit over the entire range of available data.

It is quite possible, of course, that the characteristic wave-length at 926 A.U. which determines the optical rotatory power also plays a sensible role in the ordinary dispersion of quartz. Accordingly, a dispersion formula for the ordinary index of quartz has been fitted up in which this wave-length has been included. The absorptions at 600, 1060 and 1190 A.U. have been retained, but an additional one at 926 A.U. has been introduced. The formula is

$$n_w^2 - 1 = \frac{0{\cdot}6504\,\lambda^2}{\lambda^2 - (0{\cdot}0600)^2} + \frac{0{\cdot}1000\,\lambda^2}{\lambda^2 - (0{\cdot}0926283)^2} + \frac{0{\cdot}384\,\lambda^2}{\lambda^2 - (0{\cdot}1060)^2} + \frac{0{\cdot}2228\,\lambda^2}{\lambda^2 - (0{\cdot}1190)^2} - 0{\cdot}01193\,\lambda^2. \qquad (12)$$

The agreement between the calculated and the experimental values can be seen in Table VI below.

TABLE VI

Dispersion of quartz (for explanation see text)

λ (in μ)	n_w (expt.)	n_w (calc.)
·185467	1·67578	1·67578
·214439	1·63039	1·63038
·291358	1·58098	1·58096
·340365	1·56747	1·56747
·479991	1·55012	1·55012
·58929	1·54425	1·54425
·670786	1·54147	1·54146

The accuracy of this formula is seen to be as good as any of the formulæ suggested previously.

Measurements of dn/dt have been made by Radhakrishnan (*loc. cit.*) over a wide range of temperatures for several wave-lengths. Using the data for the ordinary index, the χ's for the several dispersion frequencies occurring in Formulæ (11) and (12) have been calculated in the manner suggested by Ramachandran (1947). Since the contribution of the infra-red frequencies is small, their variations with temperature have been neglected. It is found that the data agree well when it is assumed that the extreme ultra-violet term at 600 A.U. is unaffected by temperature in both the formulæ. In Formula (11) we shall assume that the χ's for 1060 A.U. and 1190 A.U. are the same, as these two terms are very close to each other. Similarly,

in Formula (12) we shall assume that the χ's for 926, 1060 and 1190 are the same. The χ's calculated from the two formulæ are tabulated below.

TABLE VII

Temperature variation of refractive index

λ (in μ)	Temperature 50° C.			Temperature 400° C.		
	$-\frac{dn}{dt}\times 10^5$ (expt.)	$\chi\times 10^5$ Formula (11)	$\chi\times 10^5$ Formula (12)	$-\frac{dn}{dt}\times 10^5$ (expt.)	$\chi\times 10^5$ Formula (11)	$\chi\times 10^5$ Formula (12)
·2537	0·29	2·3	2·3	1·00	3·5	3·4
·3650	0·54	2·2	2·1	1·32	3·3	3·3
·5461	0·62	2·1	2·1	1·44	3·3	3·2
·5893	0·63	2·1	2·1	1·46	3·2	3·2

It will be noticed that the values of χ obtained from formulæ (11) and (12) are almost the same. It will also be seen on comparing the above table with Tables IV and V that the χ's estimated from the data on the thermal variation of refraction and the thermal variation of rotation are about the same and also increase in the same manner with rise of temperature.

6. DISCUSSION

The fact that the rotary dispersion of quartz is expressible by a formula of the type

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

ndicates that its optical activity is a result of an interaction or coupling between the successive polarisable units in the structure of the crystal excited tc similar upper energy levels, or in other words, an interaction in the nature of resonance. In their article in the *Handbuch der Physik*, Born and Goppert-Mayer (1933) arrive at the general formula

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

It will be noticed in this formula, that in addition tc terms of the Drude type, certain quadratic terms, of the same form as suggested in this paper, are also present. In view of the close agreement between the experimental values and the values calculated from the author's formula (see Table II), it appears improbable that terms of the Drude type play any observable role

in determining the optical rotatory power of quartz in the region of the visible and the ultra-violet part of the spectrum.

In conclusion, I express my sincere gratitude to Prof. Sir C. V. Raman, F.R.S., N.L., for the kind advice and the encouragement he gave me during the course of this investigation.

7. Summary

Numerous formulæ have been proposed by several authors to account for the rapid increase in the rotatory power of quartz with decrease of wavelength. These formulæ generally consist of a large number of terms both positive and negative, principally of the Drude type, involving various hypothetical characteristic frequencies. An examination of these formulæ shows that they do not possess physical justification and are arbitrary in character. In the present investigation it is shown that the data are well represented by the single term formula

$$\rho = \frac{k\lambda^2}{(\lambda^2-\lambda_0^2)^2} \text{ or } \frac{a\nu^2}{(\nu_0^2-\nu^2)^2},$$

where $k = 7{\cdot}186$ and $\lambda_0 = 0{\cdot}0926283\,\mu$. Assuming that '$a$' is a constant with temperature and ν_0 to vary with it, the thermal variation of the optical rotatory power has been quantitatively accounted for, ν_0 being found to vary in the same manner with temperature as estimated from the thermal variation of the refractive indices of quartz.

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