FARADAY ROTATION IN CALCITE

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1. Introduction

As the presence of birefringence in a solid interferes with the measurement of its magneto-optic rotation to a considerable degree, the determination of the Faraday rotation in a highly anisotropic crystal, even along the optic axis presents some experimental difficulty. In spite of its high birefringence, it was in calcite that Chauvin[1,2] verified the laws of propagation of light in a birefringent magneto-optic medium. His choice of this substance was obviously governed by the fact that perfect specimens of this crystal are easily available. Chauvin's researches have demonstrated that the practical difficulties encountered in this field of experimentation are by no means insurmountable.

The magneto-optic rotation of calcite, apart from the problem it presents in its measurement, is interesting in many other respects. Previous researches from this laboratory on the magneto-optic properties of diamond,[3] the alums and sodium chlorate[4] have indicated that the presence of strong covalent bonds in the structure has a tendency to decrease the magneto-optic anomaly factor. Calcite with its CO_3 groups would, therefore, be an ideal substance to investigate. The present paper reports the values of the Faraday rotation in calcite parallel to the optic axis and also records the values of the Verdet constant of sodium chlorate. These measurements have proved quite fruitful in that it has been possible to establish, as had been foreseen by Darwin and Watson[5] (1927) that the magneto-optic anomaly factor for various frequencies responsible for refractive dispersion may be different.

2. The Verdet Constant in Calcite

In a very good rhomb of calcite two faces were cut and polished perpendicular to the optic axis. Great care was exercised (by a series of observations under a polarisation microscope followed by grinding and polishing of the faces) to get the optic axis eaxctly normal to these planes. The softness of the crystal, and the large variety of excellent abrasive and polishing papers that are available in the market to-day, considerably facilitated this process. The crystal was then fixed to a brass gadget (Chauvin[2]) which was capable of delicately rotating it through small angles about two perpendi-

cular axes. With this arrangement the crystal was set between the poles of the electromagnet so that light from a mercury arc passed exactly along the optic axis. The divergence of the light was reduced by first rendering it parallel and then allowing it to pass through four apertures, 3 mm. in diameter, placed at intervals of 50 cm. on either side of the calcite crystal.

Measurement of the rotation was made for λ 5461 with a sugar half shade (Rayleigh[6]), first placed at the polariser end and later at the analyser end. The magnitudes of the rotation with either arrangement did not differ by more than $\frac{1}{2}\%$ showing that the birefringence and hence the missetting of the crystal was indeed small (Ramachandran and Ramaseshan[7]). The observations for λ 5893, λ 5780 and λ 5461 were made visually and those for λ 4358 and λ 4047 were made by a spectroscopic technique developed by Landau,[8] details of which have been given in an earlier publication.[4] Table I gives the Verdet constant for calcite for different wavelengths. The specimen of calcite, 8·84 mm. thick, gave a rotation of 4·28° for a field of 16540 Oersteds.

It is estimated that the absolute magnitude of the Verdet constants reported are accurate to 3% to 4%, while the errors in the relative dispersion values are less than 1%.

TABLE I

Magneto-Optic Data for Calcite

$$\gamma_1 = \gamma_2 = 0 \cdot 825, \ \gamma_3 = 0 \cdot 20.$$

λ	Verdet Constant in min./cm./Oersted		$\bar{\gamma}$
	Exp.	Calc.	
5893	0·0185	0·0184	0·488
5780	0·0193	0·0192	0·488
5461	0·0215	0·0216	0·481
4358	0·0360	0·0355	0·480
4046	0·0418	0·0420	0·467

3. The Magneto-Optic Anomaly Factor

From the measured values of the rotation and the known refractive dispersion for the ordinary ray in calcite, the magneto-optic anomaly factor $\bar{\gamma}$ was evaluated from the modified Becquerel formula

$$V = \bar{\gamma} \cdot \frac{e}{2mc^2} \lambda \cdot \frac{dn}{d\lambda} \qquad (1)$$

These values have been entered in column 4 of Table I. One notices that with decrease in wavelength, $\bar{\gamma}$ value gradually decreases from $0 \cdot 488$ to $0 \cdot 467$. As the anomaly factor is a measure of the deviation of the Zeeman splitting of the optical absorption frequency from the normal value, the variation of γ with wavelength can only signify that the anomaly factors associated with the various optical absorption frequencies must be different.

If one assumes that the oscillator strengths (*i.e.*, the transition probabilities) of the different dispersion frequencies are not affected by a magnetic field then if

$$n^2 - 1 = \frac{A_1\lambda^2}{\lambda^2 - \lambda_1^2} + \frac{A_2\lambda^2}{\lambda^2 - \lambda_2^2} \dots \dots \qquad (2)$$

then it follows[5] that

$$V = \frac{1}{n} \frac{e}{2mc^2} \left[\frac{\gamma_1 A_1 \lambda_1^2 \lambda^2}{(\lambda^2 - \lambda_1^2)^2} + \frac{\gamma_2 A_2 \lambda_2^2 \lambda^2}{(\lambda^2 - \lambda_2^2)^2} \dots \right] \qquad (3)$$

Here γ_1, γ_2, $\dots$, etc., are the anomaly factors for the absorption wavelengths λ_1, λ_2, $\dots$

In the case of calcite it is known that the refractive dispersion can be expressed by the formula

$$n_\omega^2 - 1 = \frac{0 \cdot 43257 \lambda^2}{\lambda^2 - (0 \cdot 0500)^2} + \frac{0 \cdot 8293 \lambda^2}{\lambda^2 - (0 \cdot 1000)^2} + \frac{0 \cdot 4337 \lambda^2}{\lambda^2 - (0 \cdot 1535)^2}$$

$$+ \frac{0 \cdot 61855 \lambda^2}{\lambda^2 - (6 \cdot 7)^2}$$

(Ramachandran[8])

The magneto-optic dispersion is found to fit the formula

$$V = \frac{1}{n} \frac{e}{2mc^2} \left[\frac{0 \cdot 0008922 \lambda^2}{\{\lambda^2 - (0 \cdot 0500)^2\}^2} + \frac{0 \cdot 006842 \lambda^2}{\{\lambda^2 - (0 \cdot 1000)^2\}^2} \right.$$

$$\left. + \frac{0 \cdot 002044 \lambda^2}{\{\lambda^2 - (0 \cdot 1535)^2\}^2} \right]$$

from which $\gamma_1 = \gamma_2 = 0 \cdot 825$ and $\gamma_3 = 0 \cdot 20$. *In these formulæ V is in radians and λ in microns.* Column (3) of Table I gives the values of the Verdet constant calculated using these values of the magneto-optic anomaly factors.

The agreement between the experimental and calculated values is quite satisfactory.

It is perhaps worthwhile at this stage to emphasise that the determination of γ_1, γ_2, etc., is actually by a " fitting up " process. Therefore values of the anomaly factor of any accuracy or significance can be obtained only if one has (a) an accurate dispersion formula for refractive dispersion, using preferably experimentally observed absorption frequencies and also (b) very accurate values of the magneto-optic rotation over a wide range of wavelengths. In the case of calcite none of these conditions is very well satisfied. Even so, one notices that the anomaly factor for the absorption wavelength λ 1535, which one could associate with the CO_3 group, is much less than that for the other wavelengths.

4. FARADAY ROTATION IN NACLO₃

In view of this significant result it was considered worthwhile to redetermine the magneto-optic constants of $NaClO_3$ which also contains a group having strong covalent bonds. When the measurements were made it was found that the values reported by Voigt[10] and later by Ramaseshan[4] contained a serious numerical error. The reported values were too small by a factor of two. Table II gives the Verdet constants for $NaClO_3$.

TABLE II

Magneto-Optic Data for NaClO₃

$$\gamma_1 = 0 \cdot 74 \qquad \gamma_2 = 0 \cdot 46.$$

λ	Verdet Constant in min./cm./Oersted		γ
	Exp.	Calc.	
5780	0·0179	0·0179	0·668
5461	0·0203	0·0203	0·669
4358	0·0334	0·0334	0·663
4047	0·0397	0·0397	0 660
3650	0·0508	0·0507	0·653

In this case

$$n^2 - 1 = \frac{1 \cdot 1825 \, \lambda^2}{\lambda^2 - (0 \cdot 0900)^2} + \frac{0 \cdot 07992 \, \lambda^2}{\lambda^2 - (0 \cdot 1850)^2} - 0 \cdot 00864 \, \lambda^2$$

(Chandrasekhar[11])

and

$$V = \frac{1}{n} \frac{e}{2mc^2} \left[\frac{0 \cdot 007088 \, \lambda^2}{\{\lambda^2 - (0 \cdot 0900)^2\}^2} + \frac{0 \cdot 001258 \, \lambda^2}{\{\lambda^2 - (0 \cdot 1850)^2\}^2} \right]$$

from which $\gamma_1 = 0 \cdot 74$ and $\gamma_2 = 0 \cdot 46$.

The calculated values using these constants have been entered in column (3) while column (4) gives the values of the mean anomaly factor using formula (1).

Here again one notices that the γ values for an absorption wavelength associated with a covalent group (λ 1850) is much lower than that for other wavelengths. It is, therefore, worthwhile to undertake the accurate measurement of the magneto-optic rotation, over a wide range of wavelengths for a large number of crystals. For such measurements a sensitive photo-electric polarimeter for visible and ultraviolet work has been developed by one of the authors (V.S.) and the results of the studies using it will be reported in due course.

5. SUMMARY

The Faraday rotation has been measured in calcite parallel to the optic axis for the wavelengths λ 5893, λ 5780, λ 5461, λ 4358 and λ 4047 Å. A dispersion formula for the magnetic rotation has been fitted up using the same absorption frequencies that are used in refractive dispersion. It is shown that the magneto-optic anomaly factors for the various absorption frequencies are different and the value of the anomaly factor for the absorption wavelength λ 1535, is very much lower than that for the other absorption wavelengths. ($\gamma_{1535} = 0 \cdot 20$, $\gamma_{1000} = \gamma_{500} = 0 \cdot 825$.) This paper also reports the values of the Verdet constant of $NaClO_3$ for a few wavelengths from λ 5780 to λ 3650 Å. A numerical error in the earlier measurements has been noticed and rectified here. In this case also a dispersion formula for magnetic rotation has been proposed and it is found that for this crystal $\gamma_{1850} = 0 \cdot 46$ while $\gamma_{900} = 0 \cdot 74$.

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