STRESS–OPTIC DISPERSION IN GLASSES

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WHILE examining the data acquired recently by the authors on the dispersion of the stress-optic coefficient (s.o.c.) in a series of optical glasses, it was noticed that there existed no satisfactory theory on this subject for explaining the experimental results. This communication presents the phenomenological theory of stress-optic dispersion (s.o.d.) and develops a formula for the same. The data for fused silica (Filon)[1] and those obtained for a typical light glass have been used to verify the essential features of the theory.

GENERAL THEORY

The stress-optic coefficient[2] is given by the expression $B = (n_\| - n_\perp)/p$, where $n_\|$ and $n_\perp$ are the refractive indices of the stressed isotropic solid for light polarised parallel and perpendicular to the direction of stress. The magnitude of $n_\| - n_\perp$ can be easily obtained from the dispersion formula for the substance. In general, the effect of stress on a solid would be very similar to that of either a magnetic field[3] or temperature,[4,5] *i.e.*, to affect the three parameters, N_1 the number, f_1 the oscillator strength and ν_1 the frequency of the dispersion electron. If one, therefore, starts with a dispersion formula of the Drude type

$$n^2 - 1 = \sum_1^s \frac{A_1 N_1 f_1}{\nu_1^2 - \nu^2} \qquad (1)$$

then

$$\frac{\Delta n_\|}{\Delta p} = \frac{1}{2n} \sum_1^s \left\{ - \frac{2A_1 N_1 f_1 \nu_1^2}{(\nu_1^2 - \nu^2)^2} \left[\frac{1}{\nu_1} \frac{\partial \nu_1}{\partial p} \right]_\| \right.$$
$$+ \frac{A_1 N_1 f_1}{\nu_1^2 - \nu^2} \left[\frac{1}{f_1} \frac{\partial f_1}{\partial p} \right]_\|$$
$$\left. + \frac{A_1 N_1 f_1}{\nu_1^2 - \nu^2} \left[\frac{1}{N_1} \frac{\partial N_1}{\partial p} \right]_\| \right\} \qquad (2)$$

and one also has a similar expression for $\Delta n_\perp$. If one substitutes

$$K_1' = \left[\frac{1}{\nu_1} \frac{\partial \nu_1}{\partial p} \right]_\| - \left[\frac{1}{\nu_1} \frac{\partial \nu_1}{\partial p} \right]_\perp$$

and K_1'' and K_1''' for corresponding functions for the oscillator strength and the number of oscillators then

$$B = \frac{n_\| - n_\perp}{p} = \frac{1}{2n} \sum_1^s \left\{ - \frac{2K_1' A_1 N_1 f_1 \nu_1^2}{(\nu_1^2 - \nu^2)^2} \right.$$
$$+ K_1'' \frac{A_1 N_1 f_1}{\nu_1^2 - \nu^2}$$

$$\left. + K_1''' \frac{A_1 N_1 f_1}{\nu_1^2 - \nu^2} \right\} \qquad (3)$$

Since in equation (1) the sum of the transition probabilities must necessarily be equal to unity, *i.e.*, $\sum f_1 = 1$ it follows that in equation (2) $\sum \Delta f_1 = 0$ and therefore in equation (3).

$$\sum K_1'' f_1 = 0 \qquad (4)$$

Hence if the dispersion formula (1) is written as

$$n^2 - 1 = \sum_1^s \frac{C_1 \lambda_1^2 \lambda^2}{\lambda^2 - \lambda_1^2} \qquad (5)$$

then the formula for stress-optic dispersion would be

$$B = \frac{1}{n} \sum_1^s \left[\frac{P_1 C_1 \lambda_1^2 \lambda^4}{(\lambda^2 - \lambda_1^2)^2} + \frac{Q_1 C_1 \lambda_1^2 \lambda^2}{\lambda^2 - \lambda_1^2} \right] \qquad (6)$$

where

$$\sum Q_1 C_1 = 0 \qquad (7)$$

The first term (P term) represents the effect of the frequency change and second term (Q term) gives the effect due to change in the strength of the oscillators. For testing out these formulæ it is, therefore, necessary not only to have accurate data of the stress optic coefficient of a substance over a wide range of wavelengths but also to have its refractive indices analysed into an accurate dispersion formula that makes use of experimentally observed absorption frequencies.

VERIFICATION OF THE THEORY

The substances chosen for verification of this formula are fused silica and a typical crown glass. For fused quartz an accurate dispersion formula using two experimentally observed frequencies at 1190 Å and 1060 Å and a hypothetical one at 600 Å is available.[6] A two-term dispersion formula for the glass was constructed with the absorption wavelength at λ 600 and λ 1077 which fits the experimental data on refraction to five units in the fifth place of decimals. The dispersion formulæ for the two substances are as follows:

$$n^2 - 1 = \frac{0 \cdot 1570 \lambda^2}{\lambda^2 - (0 \cdot 1190)^2} + \frac{0 \cdot 4151 \lambda^2}{\lambda^2 - (0 \cdot 1060)^2}$$
$$+ \frac{0 \cdot 5320 \lambda^2}{\lambda^2 - (0 \cdot 0600)^2} + \frac{0 \cdot 4538 \lambda^2}{\lambda^2 - (8 \cdot 84)^2}$$
$$+ \frac{1 \cdot 4460 \lambda^2}{\lambda^2 - (20 \cdot 74)^2} \quad \text{(fused silica)}.$$

$$n^2 - 1 = \frac{0 \cdot 85192 \lambda^2}{\lambda^2 - (0 \cdot 1077)^2} + \frac{0 \cdot 3935 \lambda^2}{\lambda^2 - (0 \cdot 0600)^2}$$
$$\text{(Glass No. B.S.C. 510644)}.$$

The values of the s.o.c. for fused silica were obtained from a smooth curve drawn through the various values given by Filon[1] for different wavelengths. The s.o.c.'s for the glass were obtained by a magneto-optic method[7,8] developed in this laboratory.

It was first noticed that by using Q terms alone (Eqn. 6) with the condition (7) imposed, it was not possible to fit the experimental data, indicating most clearly that there does exist a frequency change when a solid is stressed. Further it was found that if one assumed that P is the same for all the frequencies, a formula with P terms only did not satisfy the experimental data. There is no doubt therefore that both frequency and transition probability changes have to be taken into account. There are now 6 parameters to be altered and one could always by a judicious variation of these, bring about a fit between the experimental and calculated values. The following reasonable assumptions have been made so that the number of parameters to be altered is reduced considerably: (a) values P and Q for λ 1190 and λ 1060 are the same, (b) the absorption wavelength at 600 Å being due to inner levels is not split by the pressure. Using these two conditions and that given in Eqn. (7) the values of the s.o.c. for different wavelengths for fused silica and the crown glass have been calculated. It must be noticed due to condition (7) the sign of Q (600) is negative, while for the other absorption wavelengths it is positive. The observed and calculated values have been entered in Tables I and II. The agreement between them is quite satisfactory. The P and Q values for different wavelengths have also been given.

TABLE I

Substance	Wavelength in Å	Stress-optic coefficients (in brewsters)	
		Calculated	Experimental
Fused Silica	6500	3·56	3·56
	6000	3·59	3·59
	5500	3·63	3·63
	5000	3·68	3·68
	4700	3·72	3·72
Glass*	5461	2·67	2·67
	4916	2·71	2·74
	4358	2·77	2·77
	4047	2·81	2·81

* B.S.C. Type—SiO_2—69·6%; B_2O_3—6·7%; K_2O—20·5%; CaO—2·9%.

TABLE II

Values of P and Q, all in units of 10^{-13}

Substance	P_1	P_2	P_3	Q_1	Q_2	Q_3
Fused Silica	6·704	6·704	0	1·451	1·451	— ·4782
Glass	2·800	..	0	2·355	..	−1·588

One is, therefore, forced to conclude that the stress-optic dispersion arises due to both a frequency shift and a change in the oscillator strength caused by strain. For unidirectional stress, which is the case under discussion, due to opposite strains parallel and perpendicular to the stress, the absorption frequency (as in the case of a magnetic field) would split into two, one of the components being responsible for the dispersion of light polarised parallel to the direction of stress, the other for light polarised in a direction perpendicular to it. Such changes in the frequency due to stress have been conceived of by earlier workers (G. N. Ramachandran[9] and Burstein[10]) and has actually been experimentally observed in Raman effect studies by Marie and Mathieu[11] in the case of quartz. The extension of this theory to cubic and anisotropic crystals is obvious. It is also evident that this theory is not applicable to the cases where there is a realignment of molecules due to stress. Such a phenomenon is known to occur in high polymers and substances containing long chain molecules. In such cases new dispersion frequencies have to be invoked to explain the dispersion.

RELATION TO CHANGES IN POLARISABILITY

A change in the density of the substance causes a variation of the Lorentz polarisation field inside it, which in its turn alters both the frequency and the transition probability of the dispersion electron. Mueller[2,12] has shown that the stress-optic effect cannot just be considered as due to changes in the Lorentz polarisation field caused by an increase in density in one direction and a simultaneous decrease in the perpendicular direction. He has also demonstrated that as the birefringence obtained by such a hypothesis would be opposite to that observed in most substances, it is essential to assume that the strain actually alters the polarisability of the atoms. The changes in the oscillator characteristics induced by density changes would, therefore, be opposite to those due to these "strain polarisability" effects.

Many of the phenomena observed in glasses can be explained if one assumes that while the atoms that actually take part in the "network" like O, Si, Al, etc., suffer changes in the polarisability due to strain and are also affected considerably by the changes in the Lorentz field, the ions like Na, K, Pb which occupy the holes in the "network"[13] are not particularly susceptible to the former polarisability changes but are affected to a larger extent by the alterations in the density of the surrounding atoms. Hence the introduction of such ions into the silica network would tend to decrease the s.o.c. of the glass. The absorption wavelengths of many of these heavier ions are much greater than those for the network forming atoms. Hence the dispersion of the negative effect would tend to mask the dispersion arising from strain polarisability changes. One could, therefore, easily conceive of glasses with a fair percentage of heavy ions having stress–optic coefficients which practically do not show any dispersion or exhibiting even negative dispersion (i.e., the s.o.c. decreasing with decreasing wavelength). As the ratio of the heavier ions continues to increase, one should expect to find the s.o.c. actually becoming zero for a particular wavelength and then changing sign.

Many of these phenomena have been observed in glasses. It has also been noticed that most glasses show a small increase in the s.o.c. at $\lambda 4916$ and a tendency to decrease at $\lambda 3650$. Investigations are in progress to find out if these anomalies are real and if so whether they have any relationship to the fluorescent bands that are usually found in these regions.

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