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METHODS OF OPTICAL BIREFRINGENCE DETERMINATION IN LIQUID CRYSTALS FROM INTERFERENCE MEASUREMENTS

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ABSTRACT Methods for the calculation of birefringence in liquid crystals from interference measurements are compared. An empirical formula, helpful in some birefringence determinations, is proposed.

The method of interference at variable wavelength^{1,2} is one of the most convenient procedures applied in refractometric studies of liquid crystals. It consists in measuring those wavelengths at which appropriately polarized light, on traversal of a planar sample, exhibits successive maxima and minima of intensity. The intensity is maximal if the difference in optical paths between the ordinary and extraordinary waves becomes equal to an integer multiple of the light wavelength:

$$\begin{aligned}\Delta n(\lambda_N) \cdot l &= N \cdot \lambda_N \\ \Delta n(\lambda_{N+1}) \cdot l &= (N+1) \cdot \lambda_{N+1} \\ &\dots \dots \dots \\ \Delta n(\lambda_{N+k}) \cdot l &= (N+k) \cdot \lambda_{N+k}\end{aligned}\tag{1}$$

Above, $\Delta n(\lambda_N)$ is the difference in refractive index between the ordinary and extraordinary wave for the wavelength λ_N , l the thickness of the sample, and N an integer. In the set of Eqs (1), $\Delta n(\lambda)$ and N are unknowns. Hence the number of the latter is by 1 larger than that of the equa-

tions. In order to determine univocally the birefringence $\Delta n(\lambda)$ from the set of equations (1), one has to make a supplementary assumption or have available the value of Δn for one wavelength from some other measurement. In our earlier paper², guided by the consideration that the distances between successive fringes vary but slowly, we assumed Δn as equal for two successive fringes. It has been shown recently^{3,4} that the above assumption can lead to large error in spite of the circumstance that, in practice, the birefringence values for two successive maxima differ but very little. In this situation M. Laurent and R. Jorneaux³ assumed the birefringence for waves, corresponding to three successive light intensity maxima, to vary by a constant amount ε :

$$\Delta n(\lambda_{N-1}) + \varepsilon = \Delta n(\lambda_N) = \Delta n(\lambda_{N+1}) - \varepsilon. \quad (2)$$

Provided the thickness l is known, Eqs (1) and (2) permit the determination of Δn for the three wavelengths corresponding to the fringes considered. On writing $1/\lambda_N = \nu_N$ and denoting the by $\Delta \nu_1 = \nu_N - \nu_{N-1}$ and $\Delta \nu_2 = \nu_{N+1} - \nu_N$ the distances between the fringes in wave-number scale, the solution of Eqs (1) and (2) takes the form:

$$\Delta n(\nu_N) = \frac{1}{l} \cdot \frac{\Delta \nu_1 + \Delta \nu_2}{\nu_N(\Delta \nu_1 - \Delta \nu_2) + 2\Delta \nu_1 \Delta \nu_2}. \quad (3)$$

Eq. (3) contains the difference $\Delta \nu_1 - \Delta \nu_2$. This is a quantity of the order of the accuracy with which ν is measured, so that Δn determined in the way described above is charged with considerable error. The error thus incurred is especially large in the region of large wave numbers (small λ), where the distances between the fringes are small.

To enhance the accuracy of $\Delta \nu$ -determination, we plotted $\Delta \nu$ versus ν for MBBA. The graph thus obtained is found to be linear throughout the whole spectral interval studied (from 15000 to 26000 cm^{-1}). Thus, we have:

$$\Delta \nu = \alpha(\nu_0 - \nu), \quad (4)$$

where α and ν_0 are the parameters of the straight line. We checked Eq. (4) for samples of various thickness finding $\alpha \cdot l = \text{const}$. We also found a similar $\Delta\nu(\nu)$ dependence for EBBA at various temperatures; this suggests that the empirical relationship (4) is by no means fortuitous.

Fig.1 shows a typical recording of interference fringes, obtained with an MBBA sample 36 μm thick, at 22°C (curve T).

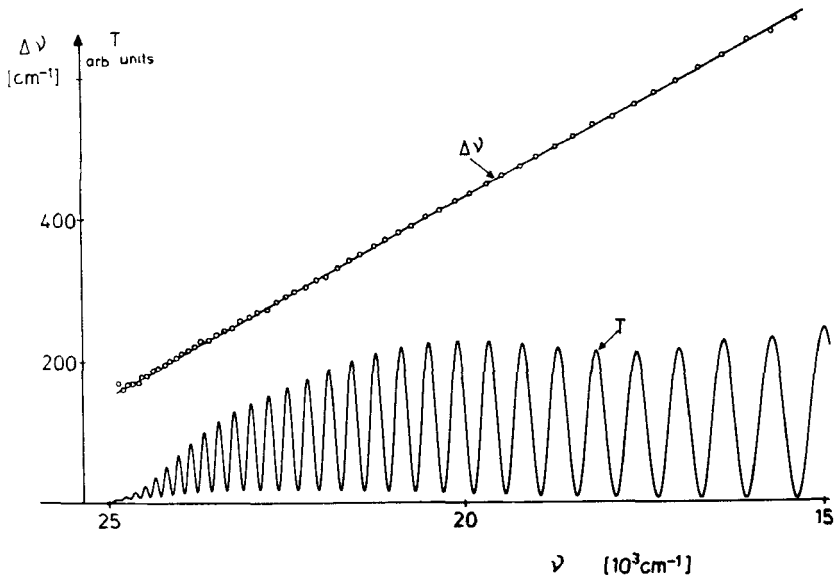


FIGURE 1. Transmission T of the sample, and distance $\Delta\nu$ between the fringes, versus the inverse light wavelength ν .

The inter-fringe distance $\Delta\nu$, determined from the curve T, is shown too. The mean square deviation of $\Delta\nu$ from the straight line is 4 cm^{-1} .

On insertion of (4) into (3), we obtain:

$$\Delta n = \frac{2}{\lambda \cdot \alpha (2\nu_0 - \nu)} \quad (5)$$

permitting the calculation of Δn for large ν

as well on the slope of the absorption band. This was not possible with Eq. (3) because of the considerable experimental error. The results of our calculations for an MBBA sample of thickness $100,6 \mu\text{m}$ at 22°C are given in Fig. 2 (curve a).

The correctness of the present results can be checked against those obtained by a different method, the accuracy of which raises no doubts. The data of Brunet-Germain⁵ obtained by the prismatic method can be taken as standard values. The method, though tedious and yielding refractive index values only for given values of the light wavelength, involves no approximations.

Comparison with the results of Brunet-Germain⁵ shows that the Δn -values obtained with Eq. (5) are too low, especially in the case of short wavelengths. Hence, the assumption (2) is still a too rough approximation.

The experimentally established linear relationship $\Delta v(v)$ permits the analytical determination of $\Delta n(v)$. Denoting by φ the difference in phase of the ordinary and extraordinary waves on traversal of the sample, we have:

$$2\pi = \varphi(v + \Delta v) - \varphi(v) \approx \frac{d\varphi}{dv} \cdot \Delta v. \quad (6)$$

On the other hand, $\varphi = 2\pi l \Delta n \cdot v$, i.e.:

$$\frac{d\varphi}{dv} = 2\pi l \frac{d}{dv} (\Delta n \cdot v). \quad (7)$$

Eqs (6) and (7) jointly yield:

$$\frac{d}{dv} (\Delta n \cdot v) = \frac{1}{l \cdot \Delta v}. \quad (8)$$

If the analytical shape of $\Delta v(v)$ is known, the differential equation (8) can be used to determine $\Delta n(v)$. For Δv given by Eq. (4), the solution of (8) is of the form:

$$\Delta n = \frac{1}{2lv} \ln \frac{C}{v_0 - v}. \quad (9)$$

Assuming Eq. (4) to be fulfilled for $v \rightarrow 0$ also,

the condition $\varphi = 0$ for $\nu = 0$ leads to $C = \nu_0$. The results of our calculations of Δn from Eq. (9) for $C = \nu_0$ are given in Fig. 2c, whence this procedure is seen to yield results more satisfactory than those derived from Eq. (5), especially for short wavelengths. Nonetheless the values of Δn are still slightly lower (by 2 - 10 %) than those of Brunet-Germain⁵. This is due to our assumption that Eq. (4) is applicable for small ν . The accuracy of Δn can be further enhanced if C is determined from a separate measurement. If C is determined using the value of Ref.⁵ for $\lambda = 643,8$ nm, Eq. (9) leads to good agreement with Brunet-Germain for the other wavelengths (Fig. 2, curve c).

Regrettably, when determining the indices n_o and n_e , the inter-fringe distance $\Delta \nu(\nu)$ cannot be described by a relationship so simple as in the case of birefringence. In this case, it is necessary to assume a value of n for a given wavelength from another, independent measurement. Chang⁴ showed how to determine the refractive indices in such cases; albeit, his procedure is by no means optimal since the experimental errors accumulate in the calculations of successive values of the indices (Fig. 2d). It appears more convenient to determine N from the equation:

$$n(\lambda_N) \cdot l = \frac{\lambda_N}{2} \cdot N, \quad (10)$$

where n is a value of the index determined from an independent measurement. Once N is available, $n(\lambda_{N+K})$ can be determined for wavelengths corresponding to the successive maxima of light intensity from the following equation:

$$n(\lambda_{N+K}) = \frac{\lambda_{N+K}}{2l} (N+K). \quad (11)$$

Since K is an integer, untainted by experimental error, the use of Eq. (11) involves no additional increase of error in the determination of n .

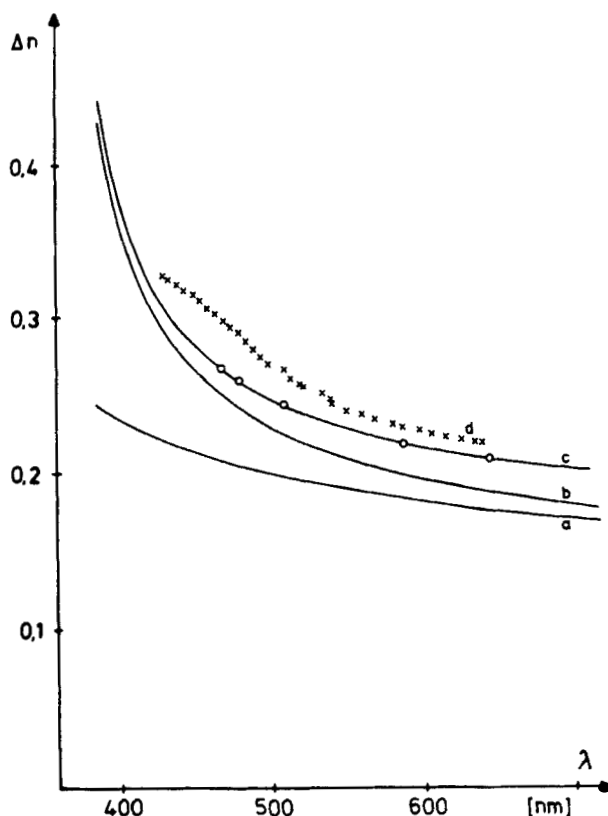


FIGURE 2. Birefringence Δn of MBBA at 22°C determined by various methods: a - from Eq. (5), b - from Eq. (9) for $C = \nu_0 = 26860\text{ cm}^{-1}$, c - from Eq. (9) for $C = 29685\text{ cm}^{-1}$ as calculated from the data of Ref.⁵ at $\lambda = 643.8\text{ nm}$, and d - with the data of Ref.⁴ (the circles denote results of Brunet-Germain⁵).

The preceding considerations lead to the conclusion the interference method at varying wavelength is not, in principle, self-sufficient. Nonetheless, it can be very useful in studies of dispersion. In cases when the empirical formula (9) proposed by us is applicable, the fun-

ctional relationship $\Delta n(\lambda)$ is accessible to determination what is very convenient especially when determining Δn near an absorption band.

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