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Wavelength and Voltage Dependences of Refractive Indices of Nematic Liquid Crystals

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Refractive indices of a mixture of nematic liquid crystals as functions both of wavelength and applied voltages were measured. The measured results are compared with a theoretical calculation in which first the distributions of tilt angle of the molecules were computed by solving a reorientation equation, then, the effective refractive indices were computed from this distribution using an index ellipsoid. It is shown that the agreement between the experimental measurements and the calculated values are fairly good within the experimental error.

1 INTRODUCTION

A determination of the refractive indices and these voltage dependences are important in the studies on the electro-optical properties of nematic liquid crystals. A number of papers were reported about the refractive indices.¹⁻¹² Among them, the measurements of the temperature dependences of the refractive indices were performed by a prism refractometer method by P. Chatelain *et al.*^{1,2} and M. Brunet-Germain.³ Recently, D. A. Balzarini⁷ and L. Bata *et al.*⁸ showed the usefulness of the utilization of laser light for determining birefringence. A few reports about only the voltage dependence have been published by H. Deuling⁹, H. Gruler *et al.*,¹⁰ L. Bata *et al.*⁸ and M. Ohtsu *et al.*,¹¹ on the other hand R. Chang¹² showed the wavelength dependence, however, the measurements of the wavelength dependence of the voltage dependent refractive indices has not been done.

In the present paper, the measured results of both the voltage and the wavelength dependences of refractive indices which were measured by the technique of interferogram^{12,13} and comparison with a theoretical calculation are described.

2 MEASURING METHOD AND EXPERIMENTAL RESULTS

The spectral intensity of transmitted light through a sample cell is measured by an ordinary spectrophotometer. The cell is composed of a pair of the parallel glass plates into which the liquid crystals are disposed. Then, an oscillatory interferogram as shown in Figure 1 is obtained. Putting the wavelengths of successive two maxima or minima of transmitted light intensity to λ_1 and λ_2 , as shown in Figure 1, one can obtain the optical length of a sample cell nd from the following equation:¹⁴

$$nd = \frac{\lambda_1 \lambda_2}{2(\lambda_2 - \lambda_1)} \quad \lambda_2 > \lambda_1, \quad (1)$$

where n is the refractive index of the medium within the gap between the glass plates. In order to determine the gap of the cell d , the measurement of

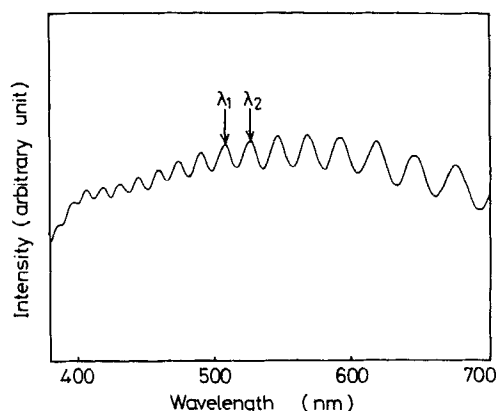


FIGURE 1 Transmitted light spectrum of a thin sample cell.

interferogram on the blank cell ($n = 1.0006$) was done. Then, the refractive indices both for the ordinary and extraordinary rays (n_o and n_e respectively) were determined from measured interferograms for the homogeneously aligned (molecules of liquid crystals are parallel to the electrodes) cell with the linearly polarized light. In measuring n_o , the optical axis of liquid crystals is set perpendicular to the electrical vector of the linearly polarized light, on the contrary n_e was measured by setting the optical axis of liquid crystals to be parallel to the electrical vector of light.

The measurements were performed on a mixture of MBBA and BBBA by 4 : 1 which has a positive dielectric anisotropy, and an AC 5 kHz fields

were applied to the cell. The temperature of the cell was regulated and fixed at 25°C by a thermoelement device.

In Figure 2 experimental results are shown as a function of a wavelength; the open and solid circles stand for the measured values of n_e and n_o , respectively. Moreover, the solid lines corresponding to n_e and n_o are calculated curves plotting the Cauchy's dispersion formula, as follows,

$$n(\lambda) = a + \frac{b}{\lambda^2} + \frac{c}{\lambda^4} \quad (2)$$

using the least-squares approximation, where λ is the wavelength and a , b and c are constant coefficients. The birefringence $\Delta n (= n_e - n_o)$ is also shown in Figure 2.

Experimental results of the voltage dependences of both n_e and Δn are shown in Figure 3 in which actual measured points are not shown but only the curves obtained by the calculation mentioned above are shown. The parameters are applied voltages to the cell in rms values.

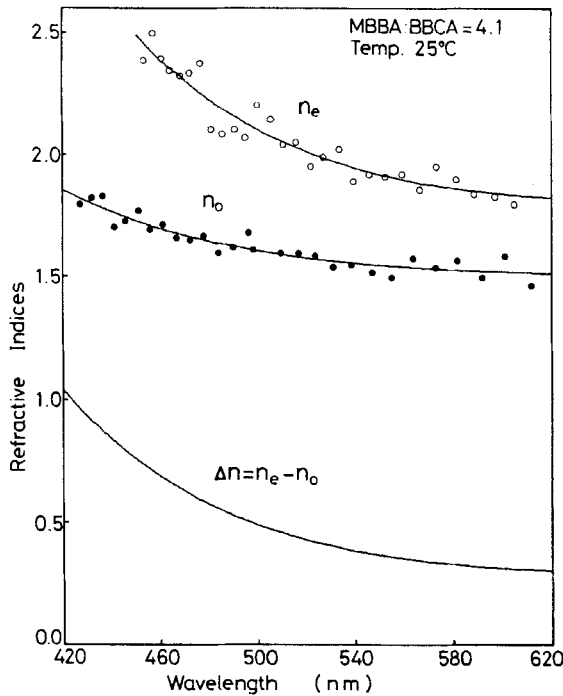


FIGURE 2 Refractive indices and anisotropy n_o , n_e and Δn as a function of a wavelength. Temperature is fixed at 25°C.

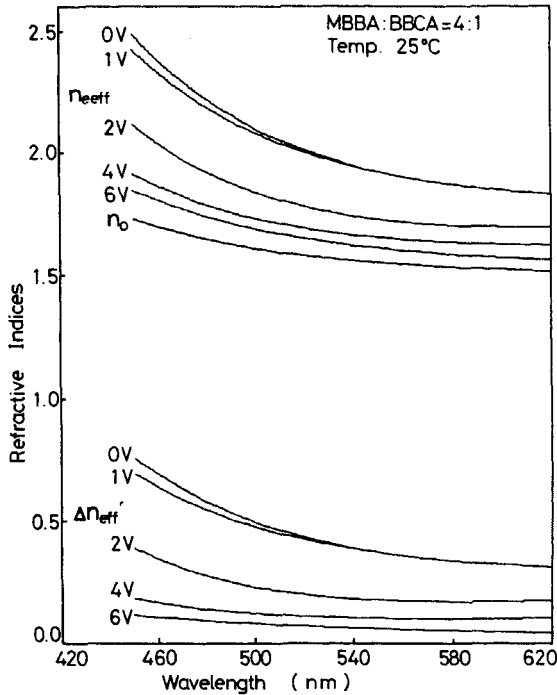


FIGURE 3 Effective refractive indices n_{eff} . A parameter is an applied voltages to the cell, an AC 5 kHz field in rms values.

3 CALCULATION OF TILT ANGLE DISTRIBUTION

In this section, it is described that the tilt angle distribution of director of nematic liquid crystals was computed in order to calculate a theoretical value of voltage dependence of the refractive index, n_{eff} .

The equations of reorientation can be derived from the Euler-Lagrange equation of the total free energy of a sample cell. For the parallel oriented sample, the elastic free energy is

$$F_{e1} = \frac{1}{2} \int_0^d (k_{11} \cos^2 \theta + k_{33} \sin^2 \theta) \left(\frac{d\theta}{dz} \right)^2 dz, \quad (3)$$

where k_{11} and k_{33} are the elastic constants, θ is the tilt angle of director and is a function of only z , and d is the thickness of the sample cell. The dielectric free energy is as follows

$$F_{\text{diel}} = \frac{1}{2} \int_0^d \mathbb{D} \cdot \mathbb{E} dz, \quad (4)$$

where \mathbb{E} is the electric field inside the sample and \mathbb{D} is the electric displacement vector. Furthermore, the vectors \mathbb{D} and \mathbb{E} are satisfied following equations

$$\begin{aligned}\text{rot } \mathbb{E} &= 0 \\ \text{div } \mathbb{D} &= 0.\end{aligned}\quad (5)$$

In our framework, it is assumed that the electric displacement is kept constant throughout the cell and that the tilt angle $\theta(z)$ will be a maximum value θ_m at $z = d/2$ and being 0 at the boundaries. Taking into account the above conditions, finally, the reorientation equations are given as follows

$$\begin{aligned}V &= 2 \left\{ \frac{k_{11}(1 + \varepsilon \sin^2 \theta_m)}{\varepsilon_0 \Delta \varepsilon} \right\}^{1/2} \int_0^{\theta_m} \left\{ \frac{1 + k \sin^2 \theta}{(1 + \varepsilon \sin^2 \theta)(\sin^2 \theta_m - \sin^2 \theta)} \right\}^{1/2} d\theta \\ Z &= \frac{d \int_0^{\theta} \left\{ \frac{(1 + k \sin^2 \theta)(1 + \varepsilon \sin^2 \theta)}{\sin^2 \theta_m - \sin^2 \theta} \right\}^{1/2} d\theta}{2 \int_0^{\theta_m} \left\{ \frac{(1 + k \sin^2 \theta)(1 + \varepsilon \sin^2 \theta)}{\sin^2 \theta_m - \sin^2 \theta} \right\}^{1/2} d\theta}\end{aligned}\quad (6)$$

where $k = (k_{33} - k_{11})/k_{11}$, $\varepsilon = (\varepsilon_{\parallel} - \varepsilon_{\perp})/\varepsilon_{\perp}$ and $\Delta \varepsilon = \varepsilon_{\parallel} - \varepsilon_{\perp}$.

Equations (6) are solved by the use of a discrete variable method as a numerical solution. The integrals in Eq. (6) are done with sufficient accuracy by a 20 point Lagrange-Gaussian quadrature. The results are presented in Figure 4. The elastic and dielectric constants used for computation are given in Table I.¹⁵

TABLE I

Physical parameters used in computation

k_{11}	6.16×10^{-7} dyne
k_{33}	7.08×10^{-7} dyne
$\Delta \varepsilon$	3.5

4 THEORETICAL VALUE OF REFRACTIVE INDEX

The voltage dependence of refractive indices for extraordinary ray, $n_{e \text{ eff}}$, are derived from the principal refractive indices, n_o and n_e , which are determined under the condition of zero field. The cell is sliced into various thin layers, and tilt angle θ is kept constant across each one layer, then $n_{e \text{ eff}}$ in these

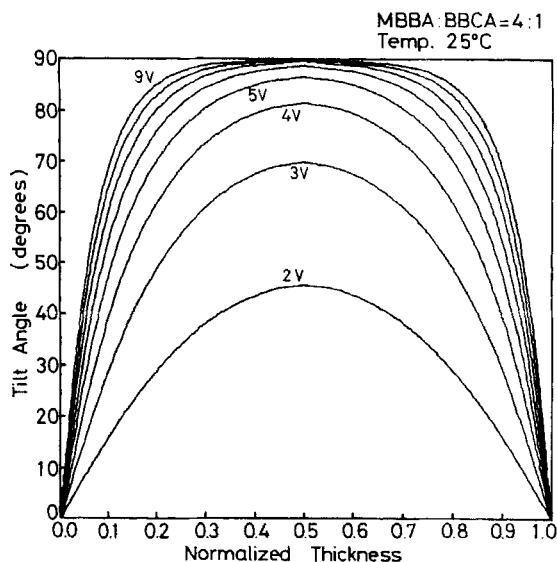


FIGURE 4 Distribution of tilt angle θ in degrees at various applied voltages as a function of Z . Temperature is fixed at 25°C.

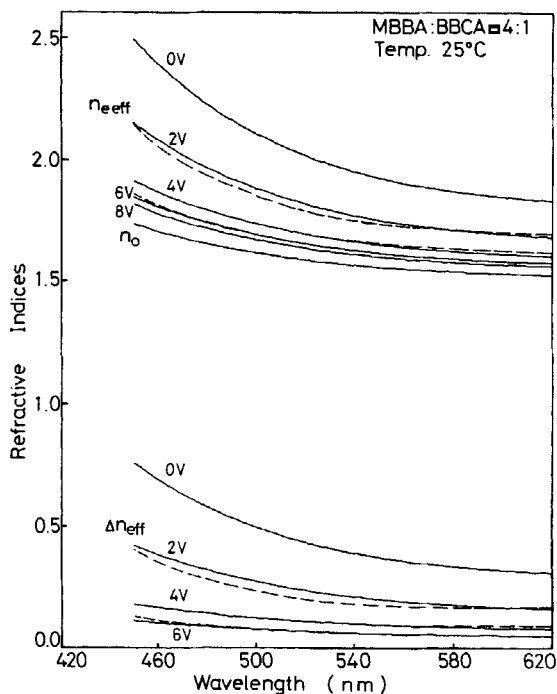


FIGURE 5 Refractive indices n_o , n_e and Δn computed from tilt angle distribution as a function of a wavelength. Solid and broken lines stand for theoretical and experimental results respectively.

layers are given by the ellipsoid given by the following equation,

$$\frac{1}{n_{\text{eff}}^2} = \frac{\sin^2 \theta}{n_o^2} + \frac{\cos^2 \theta}{n_e^2}. \quad (7)$$

Therefore, the n_{eff} is given by summing up each of the n_{eff} 's in these layers from one surface to another. In Figure 5, the computed results as a function of a wavelength are presented by solid lines. The broken lines drawn in Figure 5 are the experimental results given in the previous section (Figure 3).

The agreement between experimental and theoretical results are fairly good within the experimental error. Therefore, it is considered that this method of the calculation will be able to apply to calculate the spectral characteristics of the voltage controllable color formation.¹³

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