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# Molecular Crystals and Liquid Crystals Incorporating Nonlinear Optics

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## Magic Angle in Liquid Crystals

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#### MAGIC ANGLE IN LIQUID CRYSTALS

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Abstract Temperature dependences of the extraordinary wave refractive index and the absorption of light by dichroic dyes in oriented nematic layers have been studied. It is shown that there are such directions of the light wave relative to the nematic layer director for which the above-mentioned parameters are independent of temperature.

#### INTRODUCTION

has been shown 1,2 that in oriented nematic layers there are such directions of light wave propagation for which such important characteristics of the liquid crystal (LC) anisotropy (EW) refractive extraordinary wave index, effective absorption coefficient, optical density, optical transmission of dichroic dye in the oriented nematic, are independent of the LC temperature and have approximately the same values as they have in isotropically liquid state.

It would be of interest to generalize these results and to look at them from a slightly different point of view.

#### EXTRAORDINARY WAVE REFRACTIVE INDEX

Taking into account that the dependence of the EW refractive index on the wave propagation direction is described by the equation of ellipse

$$n_{e}(\theta) = \frac{n_{o}^{n} e}{(n_{o}^{2} \sin^{2}\theta + n_{e}^{2} \cos^{2}\theta)^{1/2}},$$
 (1)

and assuming  $n_e(\theta) = n_{NI}$  (where  $n_{NI}$  is the value of  $n_e$  for the nematic-isotropic transition), we can write the sought-for direction as follows:

$$\sin \theta_{0} = \left( \frac{(n_{NI}^{2} - n_{0}^{2})n_{e}^{2}}{(n_{e}^{2} - n_{0}^{2})n_{NI}^{2}} \right)^{1/2} . \tag{2}$$

This can be simplified assuming that  $n_{NI}^2 = (2 n_o^2 + n_e^2)/3$ . Then

$$\sin \theta_{0} \approx \frac{n_{e}^{2}}{2n_{e}^{2} + n_{o}^{2}} = \frac{n_{e}}{\sqrt{3} n_{NI}}$$
 (3)

Experimental values of the angles  $\theta_{\rm o}$  found for MBBA give the average value  $\theta_{\rm o}$  = 34.19° (Fig. 1).

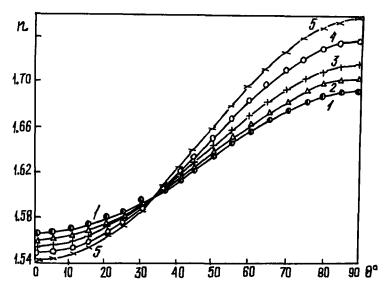


FIGURE 1 The EW refractive index as a function of the inclination angle  $\theta$  of the light beam with respect to the layer director. Curves 1-5 obtained for MBBA correspond to  $\Delta t = t_{NI} - t = 2$ , 4, 6, 14, 23°C (phase transition temperature  $t_{NI} = 45^{\circ}$ C,  $\lambda = 643.8$  nm).

If the angle is counted off from the E-vector of the light wave, then, passing to the complementary angle we obtain  $\theta_o' = 55.81^\circ$ , which is consistent within the experimental error with the value of the angle taken from the known expression describing the degree of ordering of LC molecules  $^3$ :

$$S = (3 < \cos^2 \theta > -1) / 2 \qquad , \tag{4}$$

for S = 0 ( $\theta_{\rm I}$  = 54.74°). Comparing the equations (1) and (4) for S = 0, we obtain

$$n_{e}(\theta_{I}) = n_{e}n_{o} / \left(\frac{2n_{o}^{2} + n_{e}^{2}}{3}\right)^{1/2}$$
 (5)

### ABSORPTION OF LIGHT BY DICHROIC DYE

An oriented nematic layer activated by a dichroic dye has been studied in ref.2. If we assume that, in the general case, anisotropic absorption of light of a certain frequency by a dye molecule is described by some ellipsoid of absorption and take into account that in the nematic matrix the dye molecules acquire a long-range uniaxial orientation order, then, in view of the statistical cylindrical symmetry of the molecular structure the ellipsoid of absorption will be described by ellipsoid of rotation (by analogy with the ellipsoid of refractive indices). Then, for a light wave whose E-vector oscillates in the plane coinciding with the layer director we can write:

$$k(\theta) = \left(\frac{\sin^2 \theta}{k_{\parallel}^2} + \frac{\cos^2 \theta}{k_{\parallel}^2}\right)^{1/2} , \qquad (6)$$

where  $\theta$  is the angle between the layer director and the light wave propagation direction. Substituting  $k(\theta)$  by the value of  $k_{NI}$  for the nematic-isotropic transition, we obtain:

$$\sin \theta_{0} = \left[ \frac{\left( k_{\parallel}^{2} - k_{NI}^{2} \right) k_{\perp}^{2}}{\left( k_{\parallel}^{2} - k_{\perp}^{2} \right) k_{NI}^{2}} \right]^{1/2} . \tag{7}$$

If a plane light wave falls normally to the homogeneously oriented nematic layer and the oscillation plane of the E-vector forms an angle  $\varphi$  with the layer director, then, owing to optical anisotropy of the layer two waves polarized in mutually orthogonal planes will propagate along one direction. By taking into account the absorption and anisotropy of the layer, the intensity of light passing through the nematic layer can be written as:

 $I = I_0 [\exp(-k_{\parallel}d) \cos^2 \varphi + \exp(-k_{\perp}d) \sin^2 \varphi]$ , (8) where d is the layer thickness,  $I_0$  is the incident wave intensity. Since  $I/I_0$  is the optical transmission of the layer, eqn.(8) can be simplified as

 $T(\varphi) = T_{||} + (T_{\perp} - T_{||}) \sin^2 \varphi$  . (9) Substituting  $T(\varphi)$  by the value of optical transmission in the isotropic state near the phase transition, we obtain

$$\sin \varphi_{o} = (T_{\parallel} - T_{NI})^{1/2} / (T - T_{\perp})^{1/2}$$
 (10)

As the experimental studies of various materials have shown, the angle  $\varphi_0$  = 35° with sufficient reproducibility (the error is not higher than 2%). Then, passing to the complementary angle, we again obtain 55° which coincides with the angle  $\theta_{\rm I}$  = 54.74° (see (4)). The angle  $\theta_{\rm I}$  is usually called "magic" angle. When dichroic films are studied experimentally, the value of optical transmission of the identical isotropic layer needed for calculations is measured directly on the film by rotating the polarizer into the "magic" angle.

Substituting  $\sin^2\varphi$  in eqn. (9) by  $\cos^2\varphi$  (because we passed to the complementary angle) and expressing  $\cos^2\varphi$  from (4) for S = 0, we obtain:  $T_{NI}$  = (2  $T_{||}$  +  $T_{\perp}$ )/3.

Experimental verification of eqn. (7) has revealed the stability of the absorption coefficient and optical density with changing temperature but their values differed from the calculated values more significantly. In this case, the errors near the lower mesophase boundary were as high as 5% and near the phase transition 20%, the angle being equal to 13° (the complementary angle is 77°) which is far from the "magic" angle value. A good agreement with the "magic" angle is obtained if (6) is substituted by the expression for the absorption coefficient of the extraordinary wave propagating through a uniaxial absorbing crystal. This expression is derived from the equation for the extinction coefficient 4

$$\frac{k(\theta)}{n^3(\theta)} = \frac{k_{\perp}}{n_0^3} \sin^2 \theta + \frac{k_{\parallel}}{n_e^3} \cos^2 \theta \qquad . \tag{11}$$

Then,

The average value of  $\theta_{_{\rm O}}$  in this case is equal to 36° with the error of 2% over the entire range of the mesophase.

If the layer director forms an angle  $\theta'$  with the substrate, eqn. (9) will take the form:

 $T(\varphi) = T(\theta') + (T_1 - T(\theta')) \sin^2 \varphi$ , (13) where  $T(\theta') = \exp(-D(\theta'))$ ,  $D(\theta') = k(\theta')d$  and is defined by eqn. (11). Thus, for a more general case (where the angles  $\varphi$  and  $\theta'$  change simultaneously) we obtain:

 $\sin \varphi_0 = (T_{\rm NI} - T(\theta'))^{1/2}/(T - T(\theta'))^{1/2}; \ \theta' = 90^{\circ} - \theta$ . (14) Here, we neglect the spatial divergence of o- and e-waves. Analysis of the expression obtained indicates that it is correct for  $T_{\rm NI} > T(\theta')$ . The angles  $\theta'$  for which the optical transmission T is independent of temperature will correspond to different values of  $\pm \varphi$ .

We have considered the behaviour of several parameters characterizing the anisotropy of liquid crystals along the direction corresponding to the "magic" angle. Obviously, the conclusions drawn above can be extended to include other parameters describing anisotropic properties of liquid crystals such as dielectric permittivity, heat conduction, etc. The results obtained may be useful in interpreting various nonlinear effects in liquid crystals and in choosing optimal experimental geometries.

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