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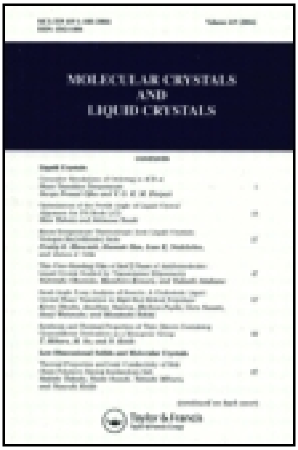

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Dispersion Relations for the Refractive Indices and the Effective Birefringence of Liquid Crystals

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Dispersion relations for the optical dielectric constants $\varepsilon_\perp$ and $\varepsilon_\parallel$ of liquid crystals are derived using Vuks' relation. These relations have three advantages over existing dispersion relations of the molecular birefringence Δn_m: 1) there is no need for having the resonance wavelength of the nearest absorption band, 2) no approximations are involved in their derivation, and 3) they are useful for calculating the effective birefringence of liquid crystals which depends on $\varepsilon_\perp$ and $\varepsilon_\parallel$ separately rather than on Δn_m. Excellent agreement is found with the available experimental results.

Keywords: liquid crystals, birefringence, dispersion, refractive indices, dielectric constants, anisotropic molecules

The birefringence dispersion of liquid crystals (LC) is important in designing full-color LC display devices and tunable filters based on LC electro-optic and magneto-optic effects. From a fundamental point of view, it is of particular interest because the birefringence is directly related to the order parameter of LC phases which exist below the isotropic phase.[1]

Birefringence dispersion in liquid crystals results from the existence of electronic absorption bands in the ultra-violet region or molecular vibrational bands in the infra-red region. Similar to the Lorentz-Lorentz[2] equation, which relates the dielectric constant to the molecular polarizability of liquids and gases at optical frequencies, Vuks[3] derived an equivalent relation for the case of anisotropic molecules. Vuks' relation is written as:

$$\frac{n_{\parallel,\perp}^2 - 1}{\langle n^2 \rangle + 2} = \frac{4\pi N}{3} \alpha_{\parallel,\perp}, \tag{1}$$

where $n_\parallel$ and $n_\perp$ are the refractive indices parallel and perpendicular to the molecular axis, respectively, $\langle n^2 \rangle = (n_\parallel^2 + 2n_\perp^2)/3$ is the average value of the optical

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dielectric constants, N is the number of molecules per unit volume and $\alpha_{\|,\perp}$ represents the molecular polarizability of the molecules parallel ($\|$) and perpendicular ($\perp$) to the molecular axis. The molecular birefringence $\Delta n_m = n_\| - n_\perp$ can be written according to Equation 1 in the form:

$$\Delta n_m = \frac{4\pi N}{3} \frac{\langle n^2 \rangle + 2}{n_\| + n_\perp} (\alpha_\| - \alpha_\perp). \tag{2}$$

Based on Equation 2 and the dispersion of $(\alpha_\| - \alpha_\perp)$, Wu[4] obtained the following dispersion relation for Δn_m:

$$\Delta n_m = G_0 \frac{\lambda^2 \lambda^{*2}}{\lambda^2 - \lambda^{*2}}. \tag{3}$$

Here, λ^* is the wavelength of the nearest electronic absorption band, G_0 is a constant which depends on the temperature and the LC mixture. The dispersion of $\alpha_{\|,\perp}$ was determined from the quantum theory of dispersion[5] in the one-band approximation and has the form:

$$\alpha_{\|,\perp} = g_{\|,\perp} \frac{\lambda^2 \lambda^{*2}}{\lambda^2 - \lambda^{*2}}, \tag{4}$$

where $g_{\|,\perp}$ are material constants which have no wavelength dependence.

The factor $\langle n^2 \rangle + 2/n_\| + n_\perp$, which appears in Equation 2, was taken in Wu's calculation[4] as a constant insensitive to the wavelength. The agreement between measured and calculated birefringence values as a function of the wavelength (Equation 3) was shown by Wu[4] to be satisfactory. The above factor was checked to be constant to within 3% for certain values of $n_\|$ and $n_\perp$. However, in general strong dispersion may exist, and the variation of this factor with wavelength must be considered, especially when high accuracy is required. Furthermore, for certain cases, such as with tilted LC structures and oblique incidence of light, one needs to evaluate the dispersion of some effective birefringence or any other function, which depends on $n_\|$ and $n_\perp$ separately rather than on Δn_m. In these cases, Equation 3 is not applicable unless additional approximations are made.

In this letter, I derive separate dispersion relations for both $n_\|$ and $n_\perp$ and for $\varepsilon_\| = n^2{}_\|$ and $\varepsilon_\perp = n_\perp^2$, starting from Vuks' relation (Equation 1) with no approximation involved. For this purpose, Vuks' relation is written in the form:

$$\frac{\varepsilon_{\|,\perp} - 1}{\varepsilon_\| + 2\varepsilon_\perp + 6} = \frac{4\pi N}{9} \alpha_{\|,\perp}. \tag{5}$$

Algebraic solution of Equation 5 yields the following:

$$\varepsilon_\perp = \frac{1 + a(7\alpha_\perp - \alpha_\|)}{1 - a(2\alpha_\perp + \alpha_\|)} \tag{6}$$

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$$\varepsilon_{\parallel} = \frac{1 + 2a(4\alpha_{\parallel} - \alpha_{\perp})}{1 - a(2\alpha_{\perp} + \alpha_{\parallel})}, \tag{7}$$

where $a = 4\pi N/9$. Using the one-band approximation, where the dispersion of $\alpha_{\parallel,\perp}$ is given in Equation 4, we obtained:

$$\varepsilon_{\perp} = \frac{G_{\perp}\lambda^2 - 1}{G\lambda^2 - 1} \tag{8}$$

$$\varepsilon_{\parallel} = \frac{G_{\parallel}\lambda^2 - 1}{G\lambda^2 - 1}, \tag{9}$$

where the constants $G_{\perp}$, G and $G_{\parallel}$ are given by:

$$G_{\perp} = \frac{1}{\lambda^{*2}} + a(7g_{\perp} - g_{\parallel})$$

$$G = \frac{1}{\lambda^{*2}} - a(2g_{\perp} + g_{\parallel})$$

$$G_{\parallel} = \frac{1}{\lambda^{*2}} + 2a(4g_{\parallel} + g_{\perp}). \tag{10}$$

These constants depend on the LC material parameters, and the temperature and they can be determined by using standard fitting procedures between measured and calculated quantities. Note that from Equations (8) and (9) the resonance wavelength λ^* need not be determined in order to calculate the dispersion, contrary to the case of Wu's[4] Equation 3 which depends directly on λ^*. The dispersion relations for the molecular birefringence Δn_m and the optical dielectric anisotropy $\delta\varepsilon = \varepsilon_{\parallel} - \varepsilon_{\perp}$ are easily found:

$$\Delta n_m = \frac{\sqrt{G_{\parallel}\lambda^2 - 1} - \sqrt{G_{\perp}\lambda^2 - 1}}{\sqrt{G\lambda^2 - 1}} \tag{11}$$

$$\delta\varepsilon = \frac{(G_{\parallel} - G_{\perp})\lambda^2}{G\lambda^2 - 1}. \tag{12}$$

As δ increases, $\varepsilon_{\perp}$ $\varepsilon_{\parallel}$, $\delta\varepsilon$ and Δn_m decreases as expected. When $\lambda >> \lambda^*$ the dispersion becomes very weak and the above quantities are determined by the constants $G_{\perp}$, G, and $G_{\parallel}$. Therefore an easy way to determine these constants is by measuring the above quantities at $\lambda >> \lambda^*$.

To illustrate the usefulness of these dispersion relations consider the case of light propagation through a homogeneously tilted LC structure with the local geometry

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as shown in Figure 1. For light propagating along the x direction and polarized in the yz plane the effective birefringence Δn_{eff} is given by:

$$\Delta n_{\text{eff}} = \sqrt{\frac{\varepsilon_{\perp}\varepsilon_{\parallel}}{\varepsilon_{\perp} + \delta\varepsilon \sin^2\theta \sin^2\varphi}} - \sqrt{\varepsilon_{\perp}}. \tag{13}$$

To calculate the dispersion of Δn_{eff} one needs the dispersion formula of $\varepsilon_{\perp}$, $\varepsilon_{\parallel}$, and $\delta\varepsilon$, but not only that for Δn_m. Equation 13 is frequently used for the case of optical switching[6] using surface stabilized ferroelectric LCs. When an electric field is applied, the azimuthal angle φ of the LC molecules varies, causing a change of the effective birefringence. Recently[7] we used Equation 13 to explain the color variations observed with the helix distortion of ferroelectric LCs. To account for the dispersion of Δn_{eff} we used the dispersion relations given in Equations 8, 9, and 12.

In Figure 2 we compare experimental results of the refractive indices $n_{\perp}$ and $n_{\parallel}$ obtained for the LC mixture BDH-E7 in its nematic phase at room temperature with the calculated values using Equations 8 and 9. These results were obtained using Talbot-Rayleigh refractometer developed recently by Warengham and Grover[8] to measure the dispersion of the refractive indices of LCs. The agreement between the measured and calculated quantities as seen in Figure 2 is good to within 1%. The constants G, $G_{\perp}$, and $G_{\parallel}$ used in the fitting are respectively: 68.5, 154.3, and 202.6 in units of μm^{-2}. A good agreement was also found between our theory and other available measurements of the refractive indices dispersion performed on different LC mixtures such as MBBA in its nematic phase,[8] Merck-1083 at different temperatures,[9] and in the ferroelectric phase of CS-1017 in the surface stabilized geometry.[10]

The generalization of Equations 8–12 to the case where many absorption bands[11] exist is straightforward by inserting the appropriate expressions for $\alpha_{\perp}$ and $\alpha_{\parallel}$ in Equations 6 and 7. In this case $\alpha_{\perp}$ and $\alpha_{\parallel}$ may be written as:

$$\alpha_{\parallel,\perp} = \sum_{i=1}^{m} g_{i\parallel,\perp} \frac{\lambda^2\lambda_i^2}{\lambda^2 - \lambda_i^2}, \tag{14}$$

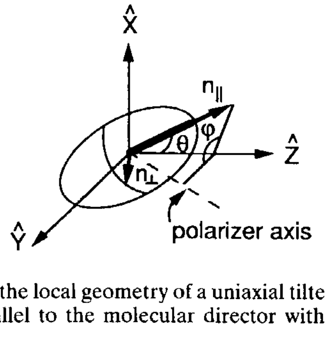

FIGURE 1 Schematic showing the local geometry of a uniaxial tilted LC structure. The index ellipsoid has its largest principal axis parallel to the molecular director with its orientation defined by the tilt angle θ and the azimuth angle φ.

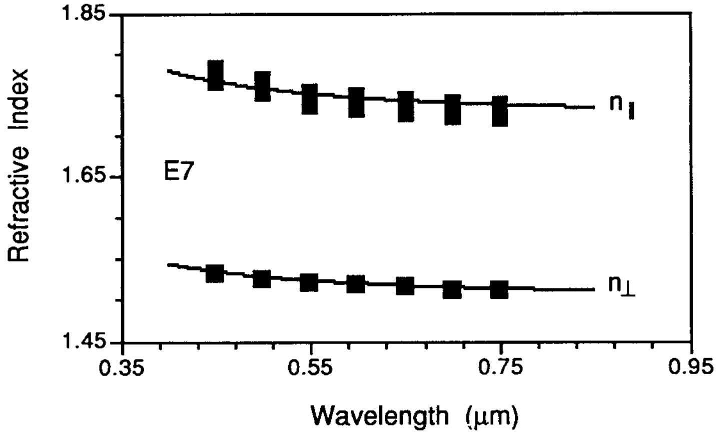

FIGURE 2 Comparison between measured (full squares and rectangles) and calculated (full curves) refractive indices as a function of wavelength for the LC mixture BDH-E7. The measured values are taken from Reference 8, and the calculations based on Equations 8 and 9.

where λ_i, $i = 1, 2, \ldots, m$, are the resonance wavelengths of the absorption bands. This case becomes increasingly complicated the greater the number of absorption bands since one needs to know all the constants $g_{i\parallel,\perp}$ and λ_i. If the absorption bands are slightly separated then the one-band approximation is adequate with λ^* being the mean resonance wavelength.

In conclusion, we have derived dispersion relations for the optical dielectric constants of LCs. These relations have several advantages over existing dispersion relations of the molecular birefringence: (1) from these relations one can easily derive dispersion relations for the optical dielectric anisotropy, the molecular birefringence and the effective birefringence; (2) in the one band approximation or when the absorption bands are slightly separated there is no need to determine the mean resonance wavelength, and (3) there is no approximation involved in their derivation. These dispersion relations are useful for designing LC light valves, tunable filters, and waveguides where control of the refractive indices and the effective birefringence of the LC is required.

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